Experimental and computational analysis of random cylinder packings with applications

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EXPERIMENTAL AND COMPUTATIONAL ANALYSIS OF RANDOM CYLINDER PACKINGS WITH APPLICATIONS

A dissertation

Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Doctor of Philosophy in

The Cain Department of Chemical Engineering

by

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NOTATION

$\phi$ = packing solid volume fraction (or packing density)
$\varepsilon$ = porosity
$L$ = particle or cylinder length
$D$ = particle or cylinder diameter
$c$ = number of random hard contacts per cylinder
$\rho$ = mean number density of a packing
$\gamma$ = the shape parameter in gamma distribution
$\beta$ = the scale parameter in gamma distribution
$\mu$ = the location parameter in gamma distribution
$Y_i^m$ = spheric harmonics
$P_i^m$ = associated Legendre polynomials
$M$ = parallel parameter of particle pair correlation
$S_{4,\alpha}$ = V-ordering parameter
$S_{2,z}, S_{4,z}$ = nematic and orthogonal correlation parameters with gravitation
$S_{2,r}, S_{4,r}$ = nematic and orthogonal correlation parameters with radial direction
$S_2, S_4, S_6$ = ordering parameters using Legendre polynomials
$I_2, I_4, I_6$ = global ordering parameters using spherical harmonics
$\xi$ = wall distance (dimensionless, normalized in the unit of cylinder diameter)
$P_c$ = capillary pressure
$P_w, P_{nw}$ = wetting phase pressure

$P_{nw}$ = nonwetting phase pressure
$\sigma$ = surface tension

$N_{\text{cap}}$ = capillary number

$P_i$ = pore pressure in a network modeling

$q_{ij}$ = volumetric flow rate between pore i and j

$G$ = hydraulic conductivity of a pore-throat

$K$ = permeability tensor

$S_i$ = wetting phase saturation in pore i

$V_i$ = volume of pore i

$R_{\text{eff}}$ = effective hydraulic radius of a pore-throat

$\psi$ = distance function

$Z$ = coordination number of a pore network
ABSTRACT

Random cylinder packings are prevalent in chemical engineering applications and they can serve as prototype models of fibrous materials and/or other particulate materials. In this research, comprehensive studies on cylinder packings were carried out by computer simulations and by experiments.

The computational studies made use of a collective rearrangement algorithm (based on a Monte Carlo technique) to generate different packing structures. 3D random packing limits were explored, and the packing structures were quantified by their positional ordering, orientational ordering, and the particle-particle contacts. Furthermore, the void space in the packings was expressed as a pore network, which retains topological and geometrical information. The significance of this approach is that any irregular continuous porous space can be approximated as a mathematically tractable pore network, thus allowing for efficient microscale flow simulation. Single-phase flow simulations were conducted, and the results were validated by calculating permeabilities.

In the experimental part of the research, a series of densification experiments were conducted on equilateral cylinders. X-ray microtomography was used to image the cylinder packs, and the particle-scale packings were reconstructed from the digital data. This numerical approach makes it possible to study detailed packing structure, packing density, the onset of ordering, and wall effects. Orthogonal ordering and layered structures were found to exist at least two characteristic diameters from the wall in cylinder packings.
Important applications for cylinder packings include multiphase flow in catalytic beds, heat transfer, bulk storage and transportation, and manufacturing of fibrous composites.
CHAPTER 1
INTRODUCTION

Random cylinder packings are of great interest because they are prototype models of many real structures, e.g. chain polymers, liquid crystals, fibrous materials and catalytic beds. These structures contribute to a wide variety of scientific research and engineering applications, including biomaterials, colloids, composites, phase transitions, fabrics, and chemical reactions. In this work, research on random cylinder packings has been carried out. The research includes: 3D analysis of the structure in random dense packings, the structural evolution of real random cylinder packs during densification, analysis of pore structure in random cylinder packings, and the simulation of fluid flow in the void space.

The first part of the work is the computer simulation of random packings of rigid cylinders, with a wide range of particle aspect ratios (1–100). A sequential parking (SP) algorithm was used to generate random loose packings and a collective rearrangement (CR) algorithm was used to generate random close packings (RCP). The latter technique is an ideal tool to reproduce disordered packing structures because of the ability to control bulk material properties (e.g. porosity, length or size distribution, and/or heterogeneity). The random dense packing limits of mono-sized cylinders in periodic domains have been explored, and results show that the maximum density decreases with the particle aspect ratio. More interesting, the existence of a weak maximum in random packing density ($\phi=0.66$ near a cylinder aspect ratio of 1.2) has been shown numerically. The phenomenon is explained by the orientational freedom gained with elongation. These simulated particulate systems might be used in different applications, including
phase transitions, liquid crystals, metastable glass, atomic structures, amorphous metals, fibrous and/or nano materials.

In the experimental part of this study, special equilateral cylinders \((L/D = 1)\) were produced and used for 3D imaging. Two sets of experiments were carried out—one in a cylindrical container with smooth surfaces and one in a cubic container with rough surfaces. The purposes of the experiments were to address the packing structures with and without wall effects during densification. A state-of-the-art X-ray microtomography system was used to obtain high-resolution images of various packings. Then, a computational algorithm was used to identify individual cylinders in the packing (e.g. their positions, orientations and sizes). Based on the particle-scale structures, the experimental packing densities of equilateral cylinders were found to fall into a range of \(0.594-0.715\), which corresponds to increasing positional and orientational ordering with increasing packing density. Statistical analysis showed that a layered structure and orthogonal ordering are two attributes found in dense cylinder packings. During the densification process, near-wall particles assume orthogonal orientations with respect to the container wall and inner particle pairs assume orthogonal orientations with respect to each other. The contact number in the densest experimental packing was found to be approximately 5.7. Detailed structural analyses (packing density, radial distribution function, local and global orientational ordering, contact number) were also performed on computer-simulated random close packings. Based on all the quantitative results, we concluded that the RCPs generated by a Monte Carlo method (collective rearrangement) do not duplicate the real dense random packings from the experiments, in which a reorganizing process was usually observed.
The void structure in cylinder packings is important in that it governs fluid flow through the packings. In chapter 5, a numerical procedure to quantify the geometry and topology of the void space in the packing is developed. The method analyzes the void structure in the framework of pore network models. In this way, the continuous void space inside a cylinder packing can be effectively expressed as a mathematically tractable, microscale pore-throat network. The resulting network represents a flow map that can be used to quantify fluid transport. Three basic elements in a network model are pores, throats, and the map of their interconnectivity. These elements correspond to larger voids, constrictions and the flow paths inside a packing. In 3D random close packings (RCPs), it was found that the normalized pore and throat sizes steadily increase as cylindrical particles become slender, and pore-throat aspect ratios maintain a value of approximately 2. The interconnectivity in the network also increases with the particle aspect ratio, which suggests a transition from tunnel-like to cage-like porous geometry inside the packings. Additional statistical results from the networks included pore size distributions (maximal inscribed diameter), throat size distributions (inscribed bottle-neck diameter), and pore-coordination-number distributions.

In chapter 6 of this work, single-phase flow simulations are described, and a set of permeability tensors from random cylinder packings are presented. In 3D random close packings, the results showed nearly isotropic permeability properties, while in two anisotropic packings (preferred horizontal or nearly nematic), anisotropic permeability properties were found. The derived permeabilities match extensive experimental data for fibrous materials, which suggests the correctness of network approach. Hence, a predictive means of computing permeability of fibrous porous media is proposed. For
multiphase flow simulations, a model formulation for dynamic imbibition and a preliminary numerical procedure were proposed, which integrate detailed microscale pore phenomena. Simulations will be conducted in the future.
CHAPTER 2
BACKGROUND AND LITERATURE REVIEW

2.1 Random Packing

The random packing structure of particles is of interest in a number of applications. Packings of simple shapes serve as prototype models for porous materials; packing structure dictates the bulk density of particulate materials; and, understanding microscale structure provides insight into properties such as thermal, fluid, and electrical conductance of various materials. Simulated packings also provide fundamental insight into molecular arrangements in crystalline and semi-crystalline materials.

2.1.1 Sphere Packing

The most widely studied packings are those composed of uniform-sized spheres. Ordered structures include rhombohedral packings and cubic packings, which bracket the range of attainable solid volume fraction (SVF) at \( \phi_{\text{max}} = 0.7405 \) and \( \phi_{\text{min}} = 0.5326 \) respectively. Disordered packings exhibit a much smaller range of SVF values. A key parameter of interest is the random-close-packed (RCP) porosity limit, which has been investigated using experiments (e.g., Scott and Kilgour, 1969; Berryman, 1983), theory (e.g., Gotoh and Finny, 1974), and numerical simulations (e.g., Jodrey and Tory, 1981). No generalized theory has provided an exact value, but the well-accepted limit is \( \phi_{\text{max}} = 0.64 \) (plus or minus some small amount that varies according to the source consulted). Generally, real packings fall into the fairly narrow density range \( \phi = 0.60 – 0.64 \) as long as the spheres are nearly uniform in size. Related studies have aimed to quantify void structure (Mellor, 1989; Nolan and Kavanagh, 1994), changes in packing
structure during densification (Clarke and Jonsson, 1993), radial distribution function (Bennett, 1972; Clark and Jonsson, 1993; Finney, 1970), contact coordination number (Bennett, 1972), and boundary effects (Reyes and Iglesia, 1991). Finally, it is worth noting that Torquato et al. (2000) have proposed the maximally random jammed (MRJ) state as a more rigorous definition than the RCP limit to characterize the dense, disordered structures.

2.1.2 Cylinder Packing

Packings of cylindrical particles are likewise of interest because of the application to real systems such as fibrous materials and catalyst beds. The most fundamental distinction between sphere and cylinder packings is that the latter systems require one additional orientational vector of each particle for the structure to be fully characterized. This extra orientational freedom leads to a number of interesting questions, e.g. is there a widely-accepted solid-volume fraction in random close packing of cylinders with a specific particle aspect ratio? What is the maximal random packing density (both experimental and simulated) of equilateral cylinders compared to the packing of spheres? How does the structure evolve when a cylinder pack undergoes a densification process? Can computer simulations imitate the densification process and generate a maximal random jammed (MRJ) structure? These interesting questions have drawn a number of scholars to examine cylinder packings, either by experiments, statistical analysis or simulations.

Fundamental experimental research on cylindrical packing was carried out since 1970s.
Milewski (1973) did detailed experiments on cylindrical packings by using wooden rods (uniform $L/D$ ratios from 4 to 72) and milled glass fibers (symmetric distribution of $L/D$). The author concluded that the cylinder aspect ratio (or average aspect ratio) determines the solid fraction of random packing, which decreases with increasing aspect ratio. The fraction from the fibers with size distribution is close to the case of rods with of uniform distribution. The work provided a series of experimental benchmarks.

Nardin (1985) generated loose and dense packings for a number of particle shapes, including fibers over large aspect ratios. It was also found that the packing volume fraction varies inversely with particle aspect ratio when the ratio is large (fibers).

Evans & Gibson (1986) used liquid-crystal theory to formulate a model for the maximum packing fraction. They proposed an analogy between the thermodynamic transition from a random to (partially) ordered state and the behavior of cylindrical packings, in which geometric considerations prohibit a random arrangement at sufficiently high values of SVF. From liquid-crystal theory, the basic formula $\phi_{\text{max}} = kD/L$ was presented, where $k$ is a constant. Theoretical computations of the probability that the rotational free volume around a fiber contains neighboring fibers gives $k = 4$. However, a fit to available experimental data gives $k = 5.3$. They suggested that those results were limited to higher particle aspect ratios of above 10.

Parkhouse & Kelly (1995) used a combination of geometric, probabilistic, and empirical arguments to obtain a general formula for the SVF of the form $\phi = 2ln(L/D)/(L/D)$. An extension to the formula was also proposed that accounted for the
possibility of filling in all available void space. That gave higher values of SVF, with an additional term of \(5\pi/(L/D)^2\).

Zou and Yu (1996) performed a series of experiments on a number of non-spherical particles in an attempt to generate loose and tight packings. They correlated their data as a function of particle sphericity \(\psi\) (defined as the ratio of the surface area of a sphere having the same volume as the particle to the surface area of the particle). For cylinders in particular, they obtained an empirical equation for the close-packed porosity limit. The limitation of their work is that no particle aspect ratio was correlated.

Philipse (1996) described the arrangement of high-aspect ratio packings from the perspective of the excluded volume available to a particle assuming a random orientation in the packing. He argued that analyzing the structure of high-aspect ratio particles is simpler than compact particles, because “for sufficiently thin rods, mechanical contacts are effectively uncorrelated: the random thin-rod array is essentially a collection of independent pairs of rods.” He suggested that \(\phi(L/D) = \langle c \rangle\) for \(L/D > 15\), where \(\langle c \rangle\) represents the number of random hard contacts per cylinder (which is constant for long particles). A value of \(\langle c \rangle = 5.4\) was proposed, thus giving essentially the same expression (a slight difference in the constant) as Evans & Gibson (1986). He pointed out that this isotropic random dense packing corresponds to metastable “rod-glasses”. He also pointed out that uncorrelated pair contacts in this model is suffice to explain densities of a variety of thin-rod systems as colloidal sediments, rheology, and percolation of random rods.

Other investigators focused on packings of smaller aspect ratio (compact) cylinders. Nardin et al. (1985) created loose and dense packings of polypropylene
For $L/D = 1$, the minimum and maximum SVF values were reported to be 0.538 and 0.629 respectively.

Dixon (1988) measured void fraction of fixed beds by using equilateral cylindrical particles. The columns were packed by fairly slow pouring of the packing by hand, without tamping or attempting to induce settling of the packing. This process suggests a random loose packing fraction of 0.64 in experiments.

Foumeny & Roshani (1991) extended Dixon’s results for equilateral cylinder ($L/D=1$) over a certain range of aspect ratios ($L/D=0.5-3$) by introducing a concept of diameter of sphere of equivalent volume. To prepare a bed, particles were simply poured into a container and gently vibrated in order to get a more settled bed. Since this process included a settling process, the resulting solid fractions are larger than the results from Dixon. Both of these authors alleged that the ratio of the equivalent cylinder diameter compared to the tube is a key factor to determine the final porosity, but they did not study the effect of aspect ratio of cylinders.

Zou & Yu (1995, 1996) proposed that the void fraction is strongly depended on the aspect ratio ($L_p/D_p$) of cylinders. They did a series of experiments on packings that include particles with aspect ratio of up to 25. Based on the experimental data, they introduced two formulas to compute RLP porosity and RCP porosity respectively.

\[
\begin{align*}
\text{Loose packing: } & \quad \varepsilon_0 = \exp \{ \psi^{5.58} \times \exp[5.89 \times (1 - \psi)] \times \ln 0.40 \} \\
\text{Close packing: } & \quad \varepsilon_\infty = \exp \{ \psi^{6.74} \times \exp[8.00 \times (1 - \psi)] \times \ln 0.36 \}
\end{align*}
\]

where \( \psi = 2.621 \times (L_p/D_p)^{2/3} / [1 + 2 \times (L_p/D_p)] \)

Computer simulations on random cylinder packings are rare compared to simulations on random sphere packings. The earliest computer work on rod-like particles ...
is from Vold (1959) who built a numerical model to simulate the sedimentation of dilute dispersions of randomly oriented rod-like particles with aspect ratio from 1 to 18. The process is similar to the random loose packing algorithm. Because of the assumption of coherence between particles, their derived solid fractions are very low. The model can apply to suspension of lithium stearate in benzene.

Evans and Ferrar (1989) performed computer simulations to generate random packings of thick fibers with aspect ratios between 1 and 30. However, they used a parking algorithm, which is known to generate volume fractions that are significantly lower than in real structures (Cooper, 1987). Additionally, they approximate each cylinder as a series of overlapping spheres along a line (to simplify calculations). This latter approximation affects the overlap calculations at the fiber ends, and has a particularly large influence on low-aspect-ratio particles.

Coelho et al. (1997) used a sequential deposition (SD) algorithm to construct packings of cylinders with aspect ratios in the range of 0.1 and 10. However, SD algorithms do not allow for good control over solid volume fraction and tend to produce stable but loose packings. This problem is evidenced by tests of their algorithm to generate sphere packings, which produced $\varepsilon = 0.402$ (significantly higher than the RCP limit).

Nandakumar et al. (1999) used computational solid-geometry and collision-detection algorithms to create packed structures with arbitrary-shaped particles. This approach is again a SD-type algorithm, and results show the same loose-packed characteristics.
Some researchers have examined other nonspherical particles (including ellipsoids, spheroids, and spherocylinders), in which the geometry of particle-particle contacts is simplified. These particles also have axis-symmetric geometry. For the packings of very large aspect ratios, all these nonspherical particles form quite similar structures. Sherwood (1997) studied the random packing of spheroids by computer simulations. Because a parking algorithm was used, the packing densities are significantly lower than the value for real systems. Williams & Philipse (2003) worked on spherocylinders. They simulated random packings of spherocylinders by a mechanical-contraction method. They found that the parameter governing the packing limit is the particle aspect ratio. As particle aspect ratio increases, the maximal packing density decreases. A new yet puzzling phenomenon is that for compact spherocylinders (e.g. low aspect ratios), there is a density hump ($\phi = 0.695$) in the maximal packing limit near aspect ratio of 0.4. Donev et al. (2004) observed a similar phenomenon in both computer-generated and real packings of ellipsoids, and highest density ($\phi = 0.735$) occurs at aspect ratio of 1.3.

2.2 Packing Structure

2.2.1 Radial Distribution Function

The radial distribution function (RDF or $g(r)$) is a way to quantify the average structure of positional ordering in a molecular system. For spherical particles, it is used to describe how the particles are radially packed around each other and have a structure of layered extension. Traditionally, it is defined as

$$g(r) = \frac{N(r)}{N_{\text{ideal}}(r)} = \frac{N(r)}{\rho \cdot V_{\text{shell}}(r)} = \frac{N(r)}{\rho \cdot 4\pi r^2 dr}$$  (2-1)
where \( \rho \) is the mean number density. The shell volume \( V_{\text{shell}} \) is simply a spherical crust that has radius \( r \) (also can be viewed as pair separation distance) and thickness \( dr \). A typical RDF plot shows a number of important features. Firstly, at short separations (small \( r \)) the RDF is zero. This indicates the effective width of the atoms, since they cannot approach any more closely. Secondly, a number of obvious peaks appear which indicates that the atoms pack around each other in shells of neighbors. The occurrence of peaks at long range indicates a high degree of positional ordering. At very long range, every RDF tends to a value of 1, which happens because the RDF describes the average density at this range. A schematic of an RDF is showed in Figure 2.1 below.

![Figure 2.1 A schematic of RDF in spherical particle system](image)

2.2.1.1 Generalization of Radial Distribution Function

To extend the RDF to cylinder systems, the pair distance is interpreted as a minimal distance from a cylinder center to the reference cylinder surface. Therefore, an effective distance between two cylinders is from 0.5 to some large number in finite domain. The mean number density \( (\rho) \) is the ratio of the total number of cylinders and the computational domain volume. The shell is a rotated crust around the reference cylinder, which includes a cylindrical part and two rotated caps (see Figure 2.2).
With periodic boundary conditions, we can treat all the cylinders in the primary domain as reference particles to obtain an improved average RDF.

\[
\bar{g}(r) = \frac{N(r)}{N_{fibers} \cdot \rho \cdot V_{shell}(r)} 
\]

(2-3)

Note that the distance of pair separation can also be interpreted as a minimal distance between two cylinder surfaces, and an effective distance will be starting from zero. A complicated excluded volume theory with statistical analysis will be involved. Interested reader may refer to the papers by (Onsager, 1949; Blaak et al., 1999; Williams, 2003).

2.2.2 Orientational Ordering Theory

2.2.2.1 Ordering in Liquid Crystal

Liquid crystals are a phase of matter whose order is intermediate between that of a liquid and that of a solid. The molecules in liquid crystals usually take a rod-like form. In a liquid phase, molecules have no intrinsic orders and display isotropic properties. In highly ordered crystalline solid phase, molecules have no translational freedom and
demonstrate anisotropic properties. That is, the molecules in solids exhibit both positional and orientational ordering; in liquids, the molecules do not have any positional or orientational ordering – the direction the molecules point and their positions are random.

![Illustration of structures of solid, liquid crystal and liquid](image)

Figure 2.3 Illustration of structures of solid, liquid crystal and liquid

There are many types of liquid crystal states, depending upon the amount of ordering in the material. These states include: (a) Nematic (molecules have no positional order but tend to point in the same direction along the director); (b) Smectic (molecules maintain the general orientational order of nematic, but also tend to align themselves in layers or planes); (c) Cholesteric (nematic mesogenic molecules containing a chiral center which produces intermolecular forces that favor alignment between molecules at a slight angle to one another); (d) Columnar (molecules in disk-like shape and stacked columns).

Three following parameters completely describe a liquid crystal structure: (a) Positional order. (b) Orientational order. (c) Bond-orientational order. Positional order refers to the extent to which an average molecule or group of molecules shows translational symmetry. Orientational order represents a measure of the tendency of the molecules to align along the director on a long-range basis. Bond-orientational order
describes a line joining the centers of nearest-neighbor molecules without requiring a regular spacing along that line (or the symmetry of molecule aggregation/cluster). In non-spherical particle systems, evidence of bond-orientational ordering is rare.

2.2.2.2 Orientational Order

In a non-thermal, non-polarized particulate system (e.g. random cylinder packing), there are no forces between particles. Structure analysis suggests that the orientational ordering is more important. We will focus on the orientational orders in the structure.

- Nematic Ordering

The nematic ordering parameter provides the simplest way to measure the amount of ordering in liquid crystals. Traditionally, this parameter has the following form

\[
S = \frac{1}{2} \left( 3 \cos^2 \theta - 1 \right) \tag{2-4}
\]

where \( \theta \) is the angle between the director (preferred direction) and the long axis of each molecule. In an isotropic liquid, the order parameter is equal to 0. For a perfect crystal, the order parameter is equal to 1. Typical values for the order parameter of a liquid crystal range between 0.3 and 0.9. To find the preferred direction, a second order tensor is used, which is defined

\[
Q_{\alpha\beta} = \frac{1}{N} \sum_{i=1}^{N} \left( \frac{3}{2} u_{i\alpha} u_{i\beta} - \frac{1}{2} \delta_{\alpha\beta} \right) \quad \text{and} \quad \alpha, \beta = x, y, z \tag{2-5}
\]

where \( u_i \) is the long axis of a molecule, \( N \) is the number of molecules, and \( \delta \) is the Kronecker delta function. \( Q_{\alpha\beta} \) is symmetric and traceless. Diagonalization of \( Q_{\alpha\beta} \) gives three eigenvalues (\( \lambda_1, \lambda_2, \lambda_3 \)), which sum to zero. The director is then given by the eigenvector associated with the largest eigenvalue.
The simple nematic ordering parameter is very sensitive in a highly ordered system (e.g. close to crystalline). However, for general disordered packings, this parameter is small.

- Cubatic Ordering

The cubatic phase is another long-range orientationally ordered phase without any positional order of the particles. In these systems, the different molecular axes of the particles align in three different orthogonal directions with equal probability of each. This elusive phase has been found in computer simulations of cut spheres (Veerman & Frenkel, 1992) and short cylinders (Blaak et al., 1999).

The formulae used to quantify the cubatic ordering include Legendre polynomials and spherical harmonics, which we will introduce in the next section.

- Other Ordering

In liquid crystals, there are plenty of symmetric structures (bcc, fcc, sc, hcp, icosahedral, star, tetrahedron, etc.) depending on the molecular shape, forces and polarized attributes. In addition, the global structures depend on the external magnetic field. Therefore, it is possible to observe countless symmetric structures microscopically in nature. In our specific cylinder-packing problem, we will focus on lower-order symmetric parameters because the packing has not demonstrated many complex structures from experimental observations.

2.2.2.3 Legendre Polynomials

The Legendre polynomials are a set of orthogonal solutions from the Legendre differential equation. They provide a means to measure structural information. One of the popular criteria is \( P_2 \) to measure nematic ordering when the system displays uniaxial
symmetric properties. When a system displays cubic symmetry, $P_4$ is a good choice. However, the Legendre polynomials are related to one angle (which usually is the angle between the director and particle axis). It cannot provide detailed 3D symmetric information in crystals compared to the spherical harmonics, in which both zenith $\theta$ and azimuth $\phi$ are used.

The first few Legendre polynomials and their plots are displayed below.

\[
P_0(x) = 1
\]

\[
P_2(x) = \frac{1}{2} (3x^2 - 1)
\]

\[
P_4(x) = \frac{1}{8} (35x^4 - 30x^2 + 3)
\]

\[
P_6(x) = \frac{1}{16} (231x^6 - 315x^4 + 105x^2 - 5)
\]

Figure 2.4   Plots of some Legendre polynomials ($x=\cos\theta$)
The Legendre polynomials are easy to understand and visualize, and for low orders (e.g. 2, 4), they have very clear physical meanings. Therefore, they are used as the correlation functions to study packing structures. Another advantage is that they can be easily adapted into other coordinates (e.g. cylindrical) to study radial distribution or wall effects.

2.2.2.4 Spherical Harmonics

Spherical harmonics analysis is based on the spherical harmonic spectroscopy that is derived from Laplace equation \((\nabla^2 U = 0)\) in spherical form. The angular portion of the solution is defined as the spherical harmonics, \(Y_l^m(\theta, \phi) = \Phi(\phi)\Theta(\theta)\). The detailed procedure of solving Laplace equation in spherical coordinate can be referred to Wolfram Mathworld.

\[
\Phi_m(\phi) = \frac{1}{\sqrt{2\pi}} e^{im\phi} 
\]
\[(2-7)\]

\[
\Theta_{lm}(\theta) = \left[ \frac{2l + 1 (l - m)!}{2 (l + m)!} \right]^{1/2} P_m^l(\cos \theta) 
\]
\[(2-8)\]

\[
Y_l^m(\theta, \phi) = \sqrt{\frac{2l + 1 (l - m)!}{4\pi (l + m)!}} P_m^l(\cos \theta)e^{im\phi} 
\]
\[(2-9)\]

where \(P_m^l\) is the associated Legendre polynomial

The spherical harmonics have important symmetric properties on \(\theta, \phi\) or both, displayed by a series of harmonic spectra. Simple algebraic manipulations of them can make more conspicuous physical meanings, e.g. rotation or separation. For example, the square of the absolute of harmonics makes it symmetric in \(\phi\), and the rotated value depends on \(\theta\) only; the square of harmonics \(Y_l^l\) tells planar symmetric information. More
detailed schematic illustrations can be found in Bourke (1990). In molecular dynamic simulations, spherical harmonics have been used in evaluating translational orders, bond-orientational orders and/or orientational orders (Nelson & Toner, 1981; Steinhardt et al., 1983; Chen et al., 1995; Blaak et al., 1999; Torquato et al., 2000).

Spherical harmonics can be used for bond-orientational order or orientational order depending on how the vector \( \vec{r} \) is defined. The former defines the vector as connecting the basis particle to its neighbors, and the latter is defined as the directions of constituent particles. Therefore, bond-orientational orders express the local symmetric information for particle clusters, or how close the system is to bcc, fcc, sc, hcp, icosahedral or other common crystal states. Orientational ordering parameters represent the information of an ensemble of particle directions. For the ideal isotropic case, the average of any harmonic should be zero except \( \bar{Q}_{00} \); therefore the first nonzero average suggests the structural information, such as \( l = 4 \) for cubatic symmetry, and \( l = 6 \) for icosahedrally oriented systems (Steinhardt, Nelson & Ronchetti, 1983).

\[
Q_{lm}(\vec{r}) = Y_{lm}(\theta(r),\phi(r)) \tag{2-10}
\]

\[
\bar{Q}_{lm} = \langle Q_{lm}(\vec{r}) \rangle = \langle Y_{lm}^{*}(\theta,\phi) \rangle \tag{2-11}
\]

Two important invariant combinations of spherical harmonics are the quadratic invariant and the third-order invariant. They are used to understand structural symmetry and are frame independent. For example, \( I_2 \) can measure uniaxial and biaxial orders, and \( I_4 \) measures uniaxial, biaxial and cubatic orders (Steinhardt et al., 1983; Blaak et al., 1999).

Quadratic invariant combinations are defined as
Third-order invariant combinations are defined as

\[ W_i = \sum_{m_1, m_2, m_3} l \times m_1, m_2, m_3 \times Q_{l m_1, m_2, m_3} \]  

(2-13)

The advantage of using spherical harmonics is that complicated structural information can be expressed as simple scalars. Because of the limited crystal structures (e.g. fcc, bcc, hcp), higher order \( l \) harmonics have limited uses in applications.

2.2.2.5 Other Correlation Formulae

Coelho et al. (1996) introduced two parameters in the paper to characterize orientational grain ordering in random packings. The first one \( Q \) correlates the particle orientation with a special axis (e.g. vertical axis, \( \mathbf{g} \)); the second one \( M \) is a pair correlation of all the particle orientations.

\[ Q = \frac{3}{4} \left\{ \left\langle \cos \left[ 2 \cos^{-1} \left( \mathbf{n} \cdot \mathbf{g} \right) \right] \right\rangle + \frac{1}{3} \right\} \quad \text{for } L/D < 1 \]  

(2-14)

\[ Q = -\frac{3}{2} \left\{ \left\langle \cos \left[ 2 \cos^{-1} \left( \mathbf{n} \cdot \mathbf{g} \right) \right] \right\rangle + \frac{1}{3} \right\} \quad \text{for } L/D > 1 \]  

(2-15)

\[ M = \frac{3}{4} \left\{ \left\langle \cos \left[ 2 \cos^{-1} \left( \mathbf{n} \cdot \mathbf{n} \right) \right] \right\rangle + \frac{1}{3} \right\} \]  

(2-16)

where \( Q = 0 \) if orientations are uniformly distributed, and \( Q = 1 \) if cylinders are in a plane. \( M = 0 \) if orientations are uncorrelated, and \( M = 1 \) if cylinders are aligned.

2.2.2.6 Local, Global and Dynamic Ordering

**Global ordering** refers to the statistical value of all particle orientations in the domain; physically it represents the macroscopic/global property. **Local ordering** refers to
the statistical value of particle orientations in a very small domain; physically it represents the microscopic/local property. Dynamic ordering is the variation in orientational ordering as the computational domain increases. It gives the evolution of ordering from local to global.

Defining a local domain in cylinder packings is difficult. The questions that arise include: Is the local computational domain spherical or another shape? Should the particle geometry be considered? How do we define neighbor pairs? How do we define the pair distance (is it equal to the minimal distance between two particle surfaces or just the distance of their centers)? Because of the angular particle surfaces, one center-to-center distance can represent many surface-to-surface distances. Practically, we adopt a criterion that two particles are neighbors as long as their minimal surface distance is shorter than the averaged diameter ($D_{\text{particle}}$).

Orientational ordering in cylinder packings is a mathematical quantification of whether all cylinders’ directions (axes) are aligned globally or locally. The global orientational ordering of cylinder packings depends on their axial directions only, but not their positions. Hence, it is possible that there are numbers of packing configurations for one value of a global orientational ordering parameter. Complete characterization of orientational ordering needs both global and local measurements.

Bond-orientational ordering is often used in expressing positional information in sphere packings, in which the constituent particles have complete symmetry. In this case, only neighboring particles are involved and the particles are expressed as the mass centers effectively. However, extending this theory to cylinder packings is limited, especially for slender cylinders.
2.2.2.7 Other Methods

- Projection and Visualization

A simple and direct method to show global ordering in packings is projection. Specifically, a vector is used to represent the axis (orientation) of a cylinder; then it is moved to the origin and normalized to the unit length, the end point expresses the orientation of the cylinder uniquely. An ensemble of them forms an image that is projected as points scattered on a unit spherical surface (Fig 2.5). If all the projected points are uniformly distributed on the unit spherical surface, it is called orientationally isotropic. Otherwise, it is called non-isotropic. However, it is a qualitative ordering measurement, detailed complex symmetric information sometimes is hard to discern by eyes.

![Figure 2.5 Projection and visualization of global ordering](image)

Liu & Thompson (2000) introduced an alignment fraction to characterize the global ordering in sphere packings. First, vectors are generated by connecting all the centers of the neighboring spheres in a packing. Then, a check is made for repeated appearance of the same vector (within some small tolerance) that appears most often. The fraction of this same vector is an indication of the global ordering. In their paper, this
fraction is set to 0.04. Therefore, a smaller alignment fraction is considered a random structure. This method can also be borrowed to evaluate orientational ordering in cylinder packings. However, the arbitrary small value chosen for random limit makes it a qualitative approach.

- Angle Distribution

In ideal isotropic packings, the angles of all particle axes form a unique distribution (we believe). Theoretically, the azimuth angles $\phi$ form a uniform distribution, and the zenith angles $\theta$ form a non-uniform distribution. If the types of distributions are known, parametric statistics will be an alternative approach.

2.3 Network Structure of Void Space

Fluid flow in porous media occurs in many applications such as synthesis of composite materials, oil and gas production, filtration, pulp and paper processing, biological transport phenomena, and design and manufacturing of adsorbent materials. Ultimately, these processes are governed by the void space in the materials. In addition, microscopic voids in atomic systems are related directly to solubility and other thermodynamic properties. To model these processes and/or to investigate fundamental physical properties, a quantitative or semi-quantitative characteristic of the void space is necessary.

2.3.1 Pore-scale Network

Due to the structural complexity and geometric irregularity, a complete characteristic of the void space in most materials is impossible. The network concept has been developed to approximate the void space by a set of pores interconnected by pore-throats. Qualitatively, pores are the larger sites in the void space; throats are the
constrictions along the flow paths connecting the pores. Thus, the continuous void phase is expressed into a pore and pore-throat network. Three basic components in a network are pores, pore-throats, and their interconnectivity, which effectively describes not only the geometry but also the topology of the void phase in porous materials. The derived network serves as a skeleton of the void phase regulating a flow process.

There are a number of advantages of using a pore-scale network approach. Firstly, it is a good compromise currently used to express any irregular pore structure without losing basic geometrical and topological properties. Secondly, pore-scale approach enables it to integrate microscopic phenomena (e.g. contact angle, capillary, film flow and snap off) in multiphase flow. Thirdly, it uses first principles of fluid mechanics to simplify flow computation (see section 2.4). Thus, it can be easily scaled up to derive macroscopic transport parameters used in the continuum-scale models.

Network structures used in network modeling are typically obtained from one of three different techniques. Most commonly, interconnected lattices are transformed into flow networks by selecting bond and node sizes from a specified size distribution. These networks are easy to generate and can reproduce certain properties of real materials in a statistical sense (Lowry & Miller, 1995; Mogensen & Stenby, 1998). A second approach is to use a simulated porous medium to construct the network. The advantage is that a simulated medium more readily captures important structural attributes and spatial correlations than a mathematical distribution. This approach also eliminates the use of scaling parameters if the true dimensionality of the model is transferred to the network. The third approach is the construction of the network from direct analysis of a real
material, e.g. SEM analysis of thin sections (Koplik et al., 1984, Ghassemzadeh et al., 2001) and microtomography (Martys, 1996, Coles, 1998).

Porescale network modeling originated from the work of Fatt (1956), who simulated granular porous media such as soil or underground rock. Later, Voronoi-Tessellation techniques were developed, through which the geometry and topology of a sphere pack were completely specified. The representative work was from Bryant et al (1993a,b), whose model predicted material permeability correctly. Another approach of extracting a network included a step of three-dimensional digital representation of pore space, where the packing models can be real media (Bakke and Oren, 1997; Bekri et al., 2000) or computer simulated packings (Hilpert et al., 2003). The models were used to study hysteretic capillary pressure-saturation relationships.

Lindquist (2001) characterized the void space of Fontainebleau sandstone using a pore network. He started from analysis of a suite of high-energy X-ray computed microtomography (CMT) images, which provide accurate measurement for the marginal and correlated distributions. Then, the medial axis transform was applied to obtain the skeleton of the void space. The pore locations, volumes, throat locations, surface area, cross sectional areas were also calculated. However, the flow model simulations from his constructed networks showed unsatisfactory residual water values ($S_w$) in both imbibition and drainage processes.

In recent years, there was a transition from stochastic network models to geologically realistic networks, in which the topologically equivalent skeleton and spatial correlation of porous medium were recorded. The transition came up with the progress of new technology, e.g. 3D X-ray microtomography. ‘Ball and Stick’ model are being
gradually discarded, pores and throats are modeled using angular (square or triangular) cross sections in favor of crevice flow dynamics (Mogensen and Stenby, 1998; Patzek, 2001; Blunt, 2001; Piri and Blunt, 2005).

2.3.2 Network in Cylinder Packings

Although network modeling has been applied to sphere packings or consolidated rocks, its use in fibrous materials is very limited. Several difficulties contribute to this limitation, e.g. anisotropic particle geometry, random structure and wide range of packing density. However, the process is still possible. The void space can be mapped by sending a ‘virtual balloon’ into the medium: the balloon would inflate and deflate as needed to fit through the pore space. The locations of balloon centers record exactly the skeleton of the medium. If pores are defined as the local maxima constrained by neighboring cylinders and throats are the maximal spheres that can pass through between two neighboring pores, the problem is not only exact in mathematics, but also reflects the real physics in multiphase flow. Without doubt, the process must be realized numerically.

Several works relating to network structure in cylinder packings are introduced below. Luchnikov et al. (1999) studied the void structures of two systems (an ensemble of straight lines and a molecular dynamics model of spherocylinders) by using generalized Voronoi-Delaunay analysis. They proposed a numerical algorithm for calculation of Voronoi network parameters, which is based on the calculation of the trajectory of an imaginary empty sphere of variable size. They qualitatively explored the distributions of the inscribed sphere radii and the bottleneck radii, and found that they were skewed. The interconnectivity was not quantitatively stated in the paper.
Ghassemzadeh et al. (2001) proposed a pore network model to study the imbibition process of paper coating. In the model, a pore network of interconnected channels between the paper’s fibers represented the void structure. They treated the interconnectivity stochastically as a parameter of the model, and studied its effects on the imbibition process. The throat sizes were chosen from a special statistical distribution, and while the pore sizes were chosen arbitrarily to be larger than the connecting throat with a random scaling factor. Their network approach was introduced in simulation of a high-speed coating process.

Thompson (2002) emphasized the scaling problems of fluid flow in fibrous materials, and predicted that the network modeling will be a standard approach in the future. In the paper, the prototype network structures were created using Voronoi diagram technique. Pore locations are the Voronoi polyhedron centers, and fibers are the edges of Voronoi polyhedron. This method facilitates the extraction of pore network process, since the pore centers are known in advance. All other network parameters can be easily calculated because the fibers were deposited in specific positions and orientations. By varying the fiber diameters, different porosity materials can be generated. However, the structures derived from the Voronoi diagrams were theoretical ones, and do not mimic any real fiber structures.

### 2.3.3 Generalized Gamma Distribution

The generalized gamma distribution is often introduced to study a skewed distribution. It is flexible enough to apply to a family of distributions because it has a shape parameter ($\gamma$), a scale parameter ($\beta$), and a location parameter ($\mu$). The PDF of this distribution is
Where $\gamma, \beta > 0$, $x \geq \mu$ and $\Gamma(\gamma)$ is the gamma function, which has the formula

$$\Gamma(\gamma) = \int_0^\infty t^{\gamma-1} \exp(-t) dt$$

Note: $E(x) = \gamma \beta$

And the CDF is defined based on PDF, which is

$$F(x, \gamma, \beta, \mu) = \int_0^x f(t, \gamma, \beta, \mu) dt$$

Because of the scale parameter, the PDF can exceed 1, so $F$ is sometimes called the mass density function. The CDF approaches 1 as $x$ becomes larger.

### 2.4 Modeling Flow in a Network

Continuum-scale modeling is usually used for fluid transport in porous materials. Continuum modeling works effectively at macroscale, but it is unable to integrate porescale behavior such as pore structure and interface instability. This disconnection (between microscopic phenomena and continuum model) tells a new way of modeling must be used, which can operate at the intermediate pore level.

Pore network model is an emerging tool for linking microscopic pore structure, flow phenomena and macroscopic properties. It can predict parametric relationships such as capillary pressure-saturation and relative permeability-saturation that are required in continuum models (Constantinides and Payatakes, 1996; Patzek, 2001; Blunt et al., 2002). Besides, it can also be used to investigate interfacial mass transfer (Dillard and Blunt, 2000), flow maldistribution (Thompson and Fogler, 1997), blob distributions and mobilization (Dias & Payatakes, 1986; Blunt & Scher, 1995), reactive flow (Fredd &
Fogler, 1998), three-phase flow (Piri & Blunt, 2005), dynamic imbibition and drainage (Mogensen & Stenby, 1998; Nguyen et al., 2004), which are widely used in reservoir flood and water aquifer remediation. Recently, there has been an increased interest in pore-scale network modeling. As Blunt (2001) summarized, the network model will be used as a platform to explore a huge range of phenomena. This multiscale approach will be cost-effective in dealing with complicated multiphase flow problems in the future.

To reflect the accurately the dynamics during multiphase flow, detailed microscale phenomena must be integrated into the model. Because of quantitative complications from film flow, snap-off, filling mechanisms, capillary pressure and detailed porous geometry, network modeling can be very challenging.

2.4.1 Microscopic Phenomena

During pore network simulation, microscopic flow phenomena occurring at the pore level must be integrated. These phenomena include: phase interface, capillarity, wettability, film flow and snap-off.

- Young-Laplace Equation

When two immiscible fluids coexist, an interface meniscus forms and a pressure difference exists between the fluids. If the displacement is quasi-statistic, the pressure difference, called the capillary pressure ($P_c$), is given by the Young-Laplace equation

$$
P_c = P_{nm} - P_w = \sigma \left( \frac{1}{R_1} + \frac{1}{R_2} \right)
$$

(2-20)

where $nw$ and $w$ label the non-wetting and wetting phases, respectively, $\sigma$ is the interfacial tension, and $R_1$ and $R_2$ are the principle radii of curvatures of the meniscus. For a cylindrical pore or throat of radius $r$, the capillary pressure is given by
where $\theta$ is the contact angle. For square capillaries, the capillary pressure is proposed by Legait (1983) and used by Mogensen & Stenby (1998)

$$P_c = \frac{2\sigma \cos(\theta)}{r} \quad (2-21)$$

$$P_c = \frac{\sigma}{r} \left( \frac{\theta + \cos^2(\theta) - \pi/4 - \sin \theta \cos \theta}{\cos \theta - \sqrt{\pi/4 - \theta + \sin \theta \cos \theta}} \right) \quad (2-22)$$

In a network, the threshold pressure (capillary pressure) of pores also depends on the number ($n$) of connected throats that are filled with non-wetting fluid (Lenormand & Zarcone, 1984). A formula was used by Blunt (1998)

$$P_c = \frac{2\sigma \cos(\theta)}{r} - \sigma \sum_{i=1}^{n} a_i x_i \quad (2-23)$$

- **Wettability**

  The wettability of a liquid to a solid is defined by its contact angle ($\theta$). If the contact angle is less than 90°, the liquid is said to be wetting to the solid; if the contact angle is greater than 90°, the liquid is nonwetting. Complete wetting means the liquid can spread along the solid surface and form a very thin liquid film (Figure 2.6).

  ![Figure 2.6 Wettability of different fluids (wetting, nonwetting, completely wetting)](image)

Wettability is important in fluid displacement. For example, in manufacturing composite materials, if the wettability between the reinforcement and fluid phase is not
satisfactory, the resulting shear strength of the composites is degraded (Yamaka, 1975; Yamamoto, 1971). Fundamental studies show that the movement of a fluid-fluid interface into a pore is strongly related to its contact angle (Lenormand et al., 1983; Li & Wardlaw, 1986; Mason & Morrow, 1994), and the void formation is related to the wettability (Dullien, 1992).

- Film Flow and Snap-off

Snap-off is believed to be the most important factor contributing to phase entrapment during displacement. For example, during imbibition at low flow rates (where the capillary pressure dominates the flow), the displacement interface can be a convex (piston-like) meniscus or a selloidal (saddle-shape) meniscus (Li & Wardlaw, 1986; Lowry & Miller, 1995). Snap-off always involves selloidal menisci. The wetting fluid flows along crevices (surface roughness such as grooves or pits or edges) in the porous space and forms a thin layer (film flow) ahead of the bulk flow front; as the layer of wetting fluid continues to swell, the radius of curvature of the fluid interface increases and the capillary pressure decreases; then, there comes a critical point where further filling of the wetting fluid will cause the interfacial curvature to decrease. At this point, the interface becomes unstable and the wetting fluid spontaneously occupies the center of porous space. This instability often ruptures the continuous nonwetting phase (Figure 2.7). Experimental observations (Lenormand et al., 1983; Li & Wardlaw, 1986) showed that snap-off could occur within throats, within pores or at the junction regions of pores and throats. In summary, snap-off is related to the competition between film flow and bulk flow, it is a complicated microscale phenomenon that depends on microscopic pore geometry and fluid properties. When it happens, the nonwetting phase becomes
disconnected and generally the process leads to the entrapment of nw phase fluid during displacement (Roof, 1970; Ng et al., 1977; Zhou & Stenby, 1993).

Figure 2.7  Schematic of snap-off (From Lenormand et al., 1983)

For a pore or throat in simple geometry of square cross section, snap-off occurs at the capillary pressure (Lenormand & Zarcone, 1984; Vidales et al., 1998; Nguyen et al., 2004)

\[
P_c = \frac{\sigma (\cos \theta - \sin \theta)}{R}
\]  

(2-24)

where \( R \) is the radius of an inscribed circumstance. For a given throat of radius \( R \), it is worth observing that this pressure is always lower than that corresponding to piston-like advance in equation above. This allows throats anywhere in the pore space to be filled with the wetting fluid in advance of the connected front. Mogensen & Stenby (1998) used a formula as the criterion for snap-off based on information about pore geometry

\[
\frac{r_{\text{throat}}}{r_{\text{pore}}} < \frac{1 - \tan(\theta) \tan(\alpha)}{2 f}
\]  

(2-25)

where \( \alpha \) is the half-angle of an angular throat and \( f \) is a geometry factor whose value is close to 1. The equation shows that it is the ratio of pore-body radius to pore-throat radius (called aspect ratio in their paper), which controls the snap-off process.
Local Capillary Pressure

As mentioned above, flow front movement can be piston-like or saddle-shaped depending on the specific wettability and local porous geometry. In either case, local capillary pressure is governed by the Young-Laplace equation. Thus, different pore-filling mechanisms correspond to specific variations in the local capillary pressure, which in turn affects the wetting pressure at the front and the whole displacement scenario. Therefore, dynamic modeling of impregnation requires detailed analysis of the variation of local capillary pressure. In the case of capillary dominated imbibition in an ideal pore, capillary pressure passes through a minimum that corresponds to the largest inscribed radius of the pore (Figure 2.8). Further increase of the wetting fluid will cause the interfacial curvature to decrease, and the capillary pressure to increase. (This phenomenon is true for any non-spherical pore geometry.) In real pores, the phenomenon is similar, but the minimum capillary pressure corresponds to a saddle-shaped meniscus. Note also that an unstable region exists for a pore in a network after the largest inscribed radius has reached (Figure 2.8), which suggests a quick filling stage so that the capillary cannot reach infinity as predicted theoretically. Therefore, in microscale modeling, local capillary pressure in a pore will a complicated function (as drafted in figure 2.9) depending on the filling mechanism, saturation, and pore geometry.

Figure 2.8 Wetting phase configuration in an idealized pore (from Thompson, 2002)
Capillary Number

Conventionally a capillary number measures the ratio of viscous to capillary forces in the system, which is defined

\[ N_{cap} = \frac{\mu u}{\sigma} \]  

(2-26)

where \( \mu \) is the viscosity of the fluid and \( u \) is the flow velocity.

2.4.2 Drainage and Imbibition

When a wetting fluid is injected into a porous medium and displaces a nonwetting fluid, the process is called imbibition; conversely, when a nonwetting fluid displaces a wetting fluid, the process is called drainage. Imbibition can be a spontaneous process where capillary forces are responsible for the fluid flow, where in drainage capillary force must be overcome by applying an external pressure drop. Often, drainage is modeled as a bond invasion-percolation process (Morgensen & Stenby, 1998; Patzek, 2001; Blunt et al., 2002). The calculations are performed in order of increasing threshold capillary entry-
pressure; only the accessible pore throats and pore bodies are invaded at each time. No entrapment of wetting phase occurs since it can always escape through wetting layers. Here we skip the details of drainage, but focus on the more complicated process of imbibition.

Compared to drainage, the microscopic picture of imbibition is more complicated. Imbibition can be slow and the invading wetting fluid can span the entire medium through corner filling. Trapping is a special case of bond-and-site invasion percolation. Patzek (2001) summarized the process as follows: when the capillary pressure decreases, the pore-throats fill in order of increasing radius, with the narrowest filling first. The throat filling starts by snap-off and then by piston-type imbibition. These two mechanisms can be described by ordinary bond percolation on a dual network and bond-invasion percolation. At the same time, the pore-bodies attached to the invaded pore-throats are subject to cooperative pore body filling by the In events. The latter mechanism and snap-off generate clusters of wetting fluid. They showed the frequency of different events (pore-throat piston-type, pore-throat snap-off, pore-body filling by In, pore-body piston-type and pore-body snap-off), which happened during imbibition simulation in the network. Blunt et al. (2002) divided the filling (imbibition) into five distinct process in waterflooding process: piston-like filling of throats, piston-like filling of pores, snap-off, forced snap-off and the collapse of oil layer, addressing the complexity of imbibition process.

Bond percolation theory describes an extreme case of imbibition during which the injection rate is infinitely low and the invading fluid is completely wetting. This implies that capillary pressure is uniform everywhere. Therefore, the invading fluid occupies the
narrowest crevices first, and then the narrower throats, and finally the pores in a wider sequence throughout the system. The capillary pressure decreases in the process as the pore space is filled in order of increasing radius (Blunt & Scher, 1995). If the displaced fluid is incompressible and the pores are much larger than throats, an unavoidable phenomenon is that all the throats are filled with the wetting fluid and the pores are filled with the nonwetting fluid. Another extreme case is at high injection rate, where the forced pressure dominates the flow. The invading fluid flows through the largest porous space with the smallest resistance. Dynamic imbibition models (which account for the full range of behavior) are much harder to construct. Qualitatively, flow rate is a primary factor controlling the competition between piston-like flow (advance through the center of pore space) and film flow (advance along crevices). At a very low flow rates, flow in crevices allows snap-off through the system and a large fraction of the nonwetting phase can be trapped. In contrast, at a higher flow rate, snap-off is suppressed, and a connected fluid front advances, trapping less nonwetting fluid.

2.4.3 Flow Simulation

Single-phase-flow simulation represents the simplest case of all network applications. The porous medium is treated as an interconnected pipe network or electrical resistor network. Specifically, pores serve as the sites for flow mixing and throats cause flow resistance. For low-Reynolds-number Newtonian flow (incompressible fluids), the flow rate of each pore-throat can be written in terms of viscosity ($\mu$), pressure difference ($\Delta P$) between nodes and a single morphologic parameter of hydraulic conductivity ($G$).
\[ q = \frac{G}{\mu} \Delta P \quad (2-27) \]

The above equation in each throat and the mass balance equation in each pore form a coupled set of linear equations, which can be used to solve the whole network system. This approach is used to compute material permeability later.

Without doubt, the hydraulic conductivity is the key term in the equation. For a circular capillary duct, a Poiseuille-type calculation is used to compute hydraulic conductivity.

\[ G = \frac{Ar^2}{8\mu L} \quad (2-28) \]

For noncircular capillary duct, Patzek & Silin (2001) derived simple analytical solutions of hydraulic conductivity based on Mason-Morrow shape factor. The duct shapes included triangular, rectangular and ellipsis. The approach can apply to throats with varied cross sections if their equivalent sizes (diameter) can be acquired. As the porosity increases, the pore geometry is evolved from tunnel-like to cage-like, and the assumptions of microscale mechanics in solving flow equation are not satisfied. The local flow picture deviates from strong axial orientation, and no equation can be used. To avoid the difficulty, Thompson (2002) proposed a novel way to compute local permeability for sparse media (e.g. fibrous material); then the local permeability was transformed into the local hydraulic conductivity. This approach is in good agreement as long as the local pore geometry is known.

\[ G = \frac{KA}{L} \quad (2-29) \]
The asymptotic expressions of local permeability were borrowed from Jackson & James (1986), which relates permeability to fiber diameter and local density. Because of the special structural generation technique of Voronoi diagram, the local porosity can be explored conveniently, however in general case, it is problematic.

The following references introduce the multiphase flow simulations using network modeling.

Blunt et al. (1992) derived relative permeabilities and capillary pressures from the simulation of two-phase flow (drainage and imbibition) in a three dimensional network. They integrated the rate-dependent competition between bulk filling and film flow in the model, and described two trapping mechanisms. They pointed out that the invasion-percolation model could not make a correct prediction for the relative permeabilities since it failed to represent the fluid connectivity. Their network model is a statistic cubic lattice (that is, ‘tubes and chambers model’ of uniform distributions of spherical pore size and cylindrical throat sizes, a fixed coordination number of six), thus the model could not represent any real physical system.

Blunt & Scher (1995) studied the two-phase flow simulations of wetting in network model. The model is again a statistic one but modifies to have angular cross section such as square. The detailed microscale physics of fluid displacement were included, which included competition between piston-like advance and flow along crevices, six dynamic pore filling mechanisms, and the perturbation effects of viscous and capillary forces. The network simulation predicted different results of finger width and residual saturation, influenced by flow patterns, which depend on flow rate ultimately.
Lowry & Miller (1995) used a network model to simulate the nonwetting-phase residual in porous media. They developed a three-dimensional random network approach of stochastic (normal, log-normal and van Genuchten pore size distributions) and variable connectivity ($Z\pm1$) to express porous media into three-dimensional networks of pores and throats. The results showed that pore-scale geometry significantly affects residual saturation, the ganglia volume distribution, and the interfacial area. However, the model could not match experimental results of ganglia volume distribution.

Mogensen & Stenby (1998) later presented a dynamic pore-scale model of imbibition. The 3D network was statistic, with a truncated Weibull type of pore size distribution. They expressed the spatial correlation of pore structure, and assigned throat radius according to the connecting pore radii. The network model was flexible, including circular, rectangular or triangular cross sections, and different interconnectivities. In the simulations, they emphasized the sensitivity analysis of capillary pressure, contact angle and aspect ratio onto the outcome of core floods, and pointed that the competition of piston-like advance and snap-off is the key dynamic for high recovery or oil entrapment.

Blunt et al. (2002) extended the network simulation further for three-phase displacement scenarios. In the paper, the authors summarized detailed pore-scale physics, and demonstrated the successful prediction of drainage and waterflood relative permeabilities for Berea sandstone. A major breakthrough is the geologically representative description of pore space, borrowed from Oren et al. (1998, 2002). The topologically equivalent network models are predictive, offering the possibility of pore-scale network modeling as a practical platform in studying a variety of transport process. (Triangular cross-sections were used in the network model.)
Nguyen et al. (2004) modeled the dynamic imbibition displacements in a network model. During simulation, the complex dynamics of frontal displacements, film swelling, snap-off and snap-off nucleated displacements were discussed. The similar conclusion of rate dependent competition (between frontal flow and snap-off) was reached. The disadvantages of the network model are the choice of regular cubic lattice and random size distribution of pores and throats.
CHAPTER 3
RANDOM CYLINDER PACKINGS

3.1 Simulated Random Cylinder Packings

3.1.1 Packing Algorithms

In this work, two numerical methods to generate random cylinder packings have been developed: a sequential parking (SP) algorithm and a collective rearrangement (CR) algorithm. Both methods can generate 3D random cylinder packings effectively. In the CR process, a certain number of cylinders (corresponding to the requirement of porosity, length and size distribution, etc.) are placed into the domain randomly. Then, a Monte Carlo perturbation process (a very small but random movement to each particle) is applied until the overlaps between all the cylinders are removed. In the SP process, cylinders are placed randomly into the domain sequentially. If a new cylinder overlaps with the old ones, it is rejected; if the new cylinder does not overlap with the old ones, it is accepted and a new cycle starts. The process is terminated with a certain stopping criterion (either a pre-set porosity or the maximum trial number). In general, a parking process generates loose structures in which the fiber particles are not physically in contact. However, a CR process can generate comparatively tighter structures, which can be viewed as gravitational stable structures with an acceptable small error. Hence, CR is more useful than the parking method. In this project, the CR method were used to model random dense packing structures. To determine the tightest packing limits, numerous CR simulations must be performed several times by trial and error. The computational algorithms are illustrated in the two flowcharts shown in Figure 3.1 & 3.2.
In put data (porosity, diameter, length, distribution etc.)

Generate a fiber particle and add into the domain randomly

Check overlap periodically

Yes

Compute new volume fraction and record fiber information

No

Terminate

Yes

No

Figure 3.1 Flowchart of the parking algorithm

Generate certain number of fiber particles and dump into the domain randomly

Apply Monte Carlo perturbation process

Check total overlap

Yes

Record fiber information

No

END

Figure 3.2 Flowchart of collective rearrangement algorithm
3.1.2 Numerical Techniques

Simulations are performed using periodic boundaries in all three Cartesian dimensions so that edge effects are eliminated and so that relatively small packings can be used in the study. For cylinders, the minimum domain size must be twice the cylinder length. Otherwise, periodicity can allow two straight cylinders to touch or overlap at two different locations, which is impossible in Euclidian space. The minimum domain size is used in many of the simulations shown below to reduce the high computational demands of the algorithm. However, larger domains are used in selected cases to ensure that results from the smaller systems are representative of general behavior. Cubic domains are used for all simulations.

Additional logic is usually added to improve performance: the random displacement direction may be weighted to encourage overlap removal, and the size of the displacements generally begins relatively large (within a cylinder radius) and becomes small as overlaps decrease. The CR algorithm has two disadvantages: it is slow and does not create hard contacts between particles. In fact, the simplistic algorithm given above generally cannot remove all overlaps within a reasonable time. A solution to this problem is to enlarge the particles slightly at first (which encourages some necessary shifting of the global structure), and then to systematically reduce particle size to the original value during convergence (Clarke and Wiley, 1987). The significant advantage of the CR algorithm, which typically outweighs the problems stated above, is the ability to specify a packing’s SVF a priori over the full range of physically possible values. Application of the CR algorithm to cylinders is somewhat more complex than to spheres because the perturbation process must account for orientation as well as location. We
have found that for smooth convergence it is important to restrict the size of the
perturbations of the cylinder orientation. Limiting these displacements in such a way that
the end of a cylinder’s axis is displaced by no more than one cylinder diameter during a
given perturbation step has proved effective in preventing cylinders from hopping across
their neighbors. Unfortunately, for large $L/D$, this restriction allows for only very small
movements of the cylinder, and therefore is a severe computational burden as the aspect
ratio becomes large.

The choice of a convergence criterion for CR algorithms varies depending on the
situation. For applied work (for instance, the modeling of fibrous materials for
engineering applications), small particle-particle overlaps are usually tolerable, and the
criterion is set so that the maximum and/or average overlaps are some small fraction of a
particle diameter. Complete overlap removal usually requires a reduction in particle
diameter (equal to the maximum overlap) at the final stage. (As described above, this
process is usually coupled with previous enlargements/reductions in particle size so that
the final diameter can be specified at the outset.) One drawback to this technique is that
the algorithm tends to converge to a point where there exist only a few overlaps of any
significance; consequently, decreasing the size of all particles by the maximum amount
(to maintain uniform particle size) gives a looser packing than what the structure
represents in a practical sense.

If the cylinder size is fixed, then SVF is determined a priori by the number of
cylinders, cylinder dimensions, and domain dimensions. Hence, determining the
maximum SVF for a given aspect ratio requires multiple simulations to be performed to
bracket a small range of SVF for which the lower value converges while the higher value
does not. In the results presented below, small but finite overlaps are allowed as a criterion for convergence. While this approach introduces some arbitrariness into the $\phi_{\text{max}}$ value, we also compute $\phi_{\text{o.f.}}$: the overlap-free SVF, which is found by reducing the cylinder radii accordingly. Thus, with confidence that $\phi_{\text{n.c.}}$ represents a non-convergent SVF, this approach produces a rigorous and reasonably small bracketed range for $\phi_{\text{max}}$.

We mention briefly the computations to detect cylinder-cylinder overlaps. This trivial calculation in the case of spheres becomes complex for cylinders and is a major factor in slowing the algorithm. The overlap computation used in this work consists of a three-step process. The first step constructs a list of closely neighboring cylinders using neighbor data compiled for the current packing configuration. The second step (applied to each pair of cylinders to be checked) is an analytical calculation to determine the perpendicular distance between the two cylinder axes, a quick check that identifies overlaps in many situations. Finally, if the step-two computation is inconclusive (because of the possibility that a cylinder end might be involved in an overlap), a numerical optimization is invoked that uses a gradient search to detect the location where a pair of cylinders most closely approach one another.

3.1.3 Isotropic Packing Results

Our computational packing results are displayed in Table 3.1. Some of the packing images are illustrated in Figure 3.3 (a)-(f).
### Table 3.1 Properties of random dense cylinder packings

<table>
<thead>
<tr>
<th>L/D</th>
<th>DDX/L</th>
<th>#Cylinders</th>
<th>φ max</th>
<th>a.o.(d)</th>
<th>max.o.(d)</th>
<th>φ o.f.</th>
<th>φ n.c.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5</td>
<td>103</td>
<td>0.6472</td>
<td>0.0002</td>
<td>0.00331</td>
<td>0.6451</td>
<td>0.6582</td>
</tr>
<tr>
<td>1</td>
<td>10</td>
<td>827</td>
<td>0.6495</td>
<td>0.0002</td>
<td>0.01214</td>
<td>0.6416</td>
<td>0.6580</td>
</tr>
<tr>
<td>1.1</td>
<td>5</td>
<td>125</td>
<td>0.6491</td>
<td>0.0002</td>
<td>0.00799</td>
<td>0.6439</td>
<td>0.6635</td>
</tr>
<tr>
<td>1.2</td>
<td>5</td>
<td>151</td>
<td>0.6589</td>
<td>0.0002</td>
<td>0.00560</td>
<td>0.6552</td>
<td>0.6741</td>
</tr>
<tr>
<td>1.5</td>
<td>5</td>
<td>230</td>
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<td>0.01536</td>
<td>0.6325</td>
<td>0.6643</td>
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<tr>
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<tr>
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<tr>
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<td>0.4243</td>
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<tr>
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<td>0.0002</td>
<td>0.02204</td>
<td>0.3418</td>
<td>0.3629</td>
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<tr>
<td>16</td>
<td>2</td>
<td>677</td>
<td>0.2596</td>
<td>0.0002</td>
<td>0.02115</td>
<td>0.2541</td>
<td>0.2674</td>
</tr>
<tr>
<td>20</td>
<td>2</td>
<td>814</td>
<td>0.1998</td>
<td>0.0002</td>
<td>0.04362</td>
<td>0.1912</td>
<td>0.2077</td>
</tr>
<tr>
<td>50</td>
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<td>350</td>
<td>0.1100</td>
<td>0.0002</td>
<td>0.01593</td>
<td>0.1083</td>
<td>0.1161</td>
</tr>
<tr>
<td>50</td>
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<td>2546</td>
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<td>0.0002</td>
<td>0.08750</td>
<td>0.0914</td>
<td>0.1071</td>
</tr>
<tr>
<td>100</td>
<td>1</td>
<td>700</td>
<td>0.0550</td>
<td>0.0002</td>
<td>0.02765</td>
<td>0.0535</td>
<td>0.0582</td>
</tr>
</tbody>
</table>

![Figure 3.3 Illustrations of compact and slender cylinder packings](image-url)  

(L/D = 1, 2, 5, 10, 20 and 50)
3.1.4 Discussion

- Compact Cylinders

For equilateral cylinders \((L/D = 1)\), the CR results from the present study give \(\phi_{\text{max}} = 0.65 \pm 0.005\). The range of values for experimental results is \(\phi = 0.6–0.71\) (Dixon, 1988; Foumeny & Roshani, 1991; Nardin et al., 1985). While the numerical result falls within this range, the maximum experimental density is significantly higher than the numerical limit. A likely explanation for this difference is the presence of local ordering in the experimental results (i.e., alignment of neighboring cylinders due to the vibration or compaction process). While no direct evidence is currently available to support this hypothesis, one additional independent data point is provided by the simulations of Coelho et al. (1997), which were performed using a sequential deposition (SD) algorithm. They report \(\phi \approx 0.55\) for a gravitationally stable packing with hard cylinder-cylinder contacts. SD algorithms are known produce lower-density packings than CR algorithms or experimental results (For sphere packings the difference is \(\phi_{\text{SD}} = 0.59\) versus \(\phi_{\text{CR}} = \phi_{\text{expt}} = 0.64\).) Given the fact that CR algorithms have effectively reproduced the dense limit for spheres and the fact that the \(\phi = 0.71\) result would require a 16% increase in density relative to Coelho et al.’s gravitationally stable density, it would appear that \(\phi_{\text{CR}} = 0.65\) is the best current estimate for the RCP limit for equilateral cylinders.

Using the \(\phi_{\text{CR}} = 0.65\) result, the issue of increasing density due to particle elongation can now be addressed. Figure 3.4(a) shows the maximum density obtained from numerical results over a relatively small range of particle aspect ratio, with results
from a number of other studies (different particle shapes) included for comparison. The data contain a small hump corresponding to a maximum density at $L/D \approx 1.2$. Although the density increase is small (in fact on the order of the error bars in the plot), the effect is real. Multiple CR simulations were performed using $L/D = 1$ and $\phi_{\text{final}} = 0.66$, none of which converged. Whereas, increasing the cylinder aspect ratio slightly to $L/D = 1.2$ allows for rapid convergence to $\phi = 0.66$ in repeated independent simulations. This observation leads to an important conclusion: while the addition of orientational freedom appears to be the dominant factor associated with increasing packing density (as in the cases of spherocylinders or ellipsoids), there exists an independent effect that stems from elongation alone, evidenced by the fact that cylinders of any aspect ratio have orientational freedom.

A second interesting observation comes from comparing the magnitudes of the densities for cylinders versus the spherocylinders and ellipsoids. The peak density for the rigid cylinders is significantly lower than the peak density for the smoother particles. However, the curves converge quite closely at unit aspect ratio (64% for spheres versus 65% for cylinders). We speculate that, in general, the sharp geometry of the cylinders leads to additional void volume in truly random packings, thus producing the 7% difference in peak densities. However, as the two particle classes approach unit aspect ratio, the loss of orientational freedom in the spheres leads to a rather dramatic drop in density, whereas little effect is observed in the cylinders because they retain orientational freedom. These results serve to clarify the suggestion made by Weitz (2004) that the density of sphere packings is anomalously low: the anomaly is not so much the density
itself (which is very similar to the cylinder packings seen here), but rather the large reduction in density as compared to the sphere’s slightly elongated cousins.

Figure 3.4  Plots of Compact and slender cylinder packing limits

- **Slender Cylinders**

  For slender particles, $\phi_{\text{max}}$ depends strongly on the aspect ratio $L/D$. This general behavior has been attributed to the fact that the average number of cylinder-cylinder contacts (which constrain movement) varies only slightly as aspect ratio increases from unity to very large values (Philipse, 1996). Despite this phenomenological explanation and data to support it, experimental and computational values for density vary widely.

  Figure 3.4(b) summarizes the experimental, theoretical, and numerical studies and illustrates the discrepancies in SVF predictions (and, to a lesser extent, discrepancies in the SVF functionality with aspect ratio). In Figure 4, computational results from the current study are added to selected results from the previously shown data. The results at high SVF agree well with the equation

$$\phi = 5.4(D/L) \quad (3-1)$$
which is the form presented by both Evans and Gibson (1986) and Philipse (1996) (ignoring the small discrepancy in the constant between these two papers). However, at moderate aspect ratios (between 10 and 40), the computed density limits are lower than Equation (3-1) predicts. In particular, consider \( L/D = 20 \): the CR algorithm predicts \( \phi_{\text{max}} = 0.20 \) whereas Equation (3-1) predicts a value of 0.27. Evans and Gibson (1986) suggest that Equation (3-1) be used for \( L/D > 10 \) and Philipse (1996) suggests that \( L/D > 15 \) is required. However, Philipse also points out that, strictly, the simple form of equation (3-1) is valid only for \( L/D \gg 1 \), since it is the asymptotic limit of the more general random-contact model. The complete equation for the special case of spherocylinders is (Philipse, 1996)

\[
\phi = \frac{L}{D} \left[ 4 \frac{D}{L} + \frac{3(L/D)}{3(L/D) + 2} \right] = \langle c \rangle
\]  

(3-2)

This equation is valid for lower aspect ratios (than Equation 1), but still requires sufficiently high aspect ratios such that particle-particle contacts remain uncorrelated. As shown in Figure 3.4, this equation provides somewhat better agreement with the computational results than the limiting form does. However, to provide a close match in the range \( \phi = 10-40 \), a value \( \langle c \rangle \approx 4.8 \) must be used. Although this functionality is in closer agreement with theories (Evans and Gibson, 1986; Philipse, 1996; Philipse and Verberkmoes, 1997), it displaces the curve in the high L/D region, which is where experiment, empirical formulations, and simulations agree well.

Comparing to experimental data we see that, in the range \( L/D \approx 10-30 \), the experiments match Equations 3-1 and 3-2 more closely than they do the CR results. However, all of the significant factors that affect experimental packings would tend to
shift the SVF to higher values. The most important of these is gravity, which tends to align the cylinders in the horizontal plane. Cylinder pairs neighboring tend to align themselves up. To a lesser extent, particle deformation and size distribution can also increase SVF. Hence, it would not be unexpected to observe experimental densities that are slightly higher than theory or computational measurements.

The inability to produce denser packings using the CR algorithm is not sufficient to prove that the higher experimental values do not represent random structures – it is certainly possible that the CR algorithm has limitations that have not been observed previously. However, this possibility must be evaluated against the experimental pitfalls and the numerous factors pointing to the CR algorithm’s validity: its success in simulating dense random sphere packings, its exact reproduction of the high $L/D$ results for cylinders, and the independent data provided by the SD simulations of Coelho et al. (1997) for the compact cylinders (discussed above). Assuming that a smooth transition to partially ordered packings does indeed occur with cylinders, this fact would make experimental determination of $\phi_{\text{max}}$ very difficult, and computational results may therefore produce the best available estimates of random packing limits.

We conclude by briefly mentioning the number of contacts per cylinder in the packings, which is an important parameter that affects thermal conductivity, strength of materials, and is a critical element of the theory proposed by Philipse (1996). Unfortunately, because the CR algorithm does not ensure hard contacts, this calculation cannot be made precisely. Estimates of hard contacts can be made by prescribing a certain minimum separation distance as equivalent to a contact. However, selection of this distance is somewhat arbitrary, and tests show that the computed number of contacts
is very sensitive to the selected separation tolerance. As an initial step, we performed the reverse calculation: i.e., determine the equivalent contact distance necessary to produce the theoretical value $\langle \gamma \rangle = 2 \cdot \langle c \rangle = 10.8$. For $L/D = 50$, this required that any cylinder-cylinder gap of $0.0086D$ or smaller be considered as a hard contact. Using this value, the distribution of particle contacts can be computed, which is significant because it cannot be measured easily in physical experiments. For the CR packing, this distribution is quite broad (i.e., a few fibers with 1 contacts, and a maximum of 22 contacts for one fiber for the $L/D = 50$ packing). However, since this behavior is partly a nonphysical consequence of the CR algorithm (which does not produce gravitationally stable packings), further work must be done to determine what aspects of these contact distributions are physically representative.

3.1.5 Anisotropic packing Results

To provide more complete data for applications, we extend our simulations to anisotropic packings: close layered and near nematic. The first structure is intended to simulate fiber mats, and the second one represents a simple liquid crystal. These packings are used to compare with the results from isotropic packings, as well as to provide a set of structural benchmarks in quantifying orientational ordering.

The simulation process is same as the one used to generate isotropic packings, except for restrictions on the initial cylinder orientations. To generate layered packings, the third orientational components are restricted to a small value (e.g. $z$ component $< 10\%$ of length). To generate nematic packings, the third orientational components are restricted to a larger value (e.g. $z$ component $> 90\%$ of length). During the Monte Carlo perturbations, there are no extra restrictions.
A series of anisotropic packings with different densities have been successfully generated. The densest isotropic packing of aspect ratio 10 is about 35%; with anisotropic packings, the densities can easily go up. Some images of anisotropic packings are illustrated in Figure 3.5. Currently, we are not trying to explore the packing limits, as real fibers in the mats are woven and curved periodically.

![Figure 3.5 Illustrations of different anisotropic packings](image)

(a. nematic 35%; b. nematic 45%; c. layered 35%; d. layered 40%)

An interesting phenomenon is that as the packing density increases, there is a higher frequency of parallel fiber bundles, even though the global structure remains random in one-dimension (nematic) or two-dimensions (layered). These fiber bundles correspond to crystal nuclei during phase transition. However, whether or not the global crystalline structure can be formed by using collective rearrangement is still unclear. The phase diagram in the dense region is quite complex: as the cubatic phase was investigated

3.2 Experimental Packs of Equilateral Cylinders

A series of packing experiments of equilateral cylindrical particles have been carried out. The experiments serve several objectives: real cylinder packs can be used to compute packing density, contact number and statistical ordering, thus providing a counterpart to the computer-simulated packings. Furthermore, two sets of experiments (one in a cubic container with rough walls and one in a cylindrical container with smooth walls) were designed, through which detailed structural information, dynamic structural evolution during the densification process, and wall effects were studied.

3.2.1 Experimental Methods

- Cylinders

The cylinders used in the experiments were constructed with an outer layer of PEEK polymer and an inner aluminum (AL) core. This choice of materials was made so that the cylinder axis can be identified easily in the microtomography images, allowing for easy identification of cylinder orientations.

The cylinders were made using PEEK tubing (Upchurch scientific), with 1.8 mm OD and 0.030” ID. A 0.025” inch OD aluminum welding rod was coated with craft adhesive and fed into the center of the tubing. The tubing was then cut into 1.8 mm long sections using an electronic tubing cutter (Figure 3.6). A feed groove was mounted into the tubing cutter along with a stop/release that allowed the tubing to be fed to the proper position before cutting. This arrangement made the cutting process relatively accurate and automated, which was necessary because of the large number of cylinders needed.
The dimensions of 40 of the cut cylinders were measured, giving the following results: $D = 1.77 \pm 0.0021$ mm; $L = 1.80 \pm 0.0024$ mm (where the ± range is the standard deviation in the measurement).

Figure 3.6  The electric tubing cutter and sample cylinders

• X-Ray Microtomography

The X-ray microtomography (XMT) imaging was performed at the Naval Research Laboratory at Stennis Space Center (Mississippi) under the guidance of Dr. Allen Reed. The XMT system is composed of an x-ray tube (i.e., FeinFocus 10-225 keV, 0-1 mA with 3 µm x-ray aperture), a 23-cm diameter image intensifier, a sample stage that provides 6-fold axes of movement, and a computer that collects and converts x-ray attenuation data into images (see Figure 3.7). The x-ray production occurs in a lead-shielded room, while the conversion of attenuation data into images occurs outside the lead-shielded room, in the control room. Also, within the control room, a video projection of a two-dimensional x-ray image is displayed onto a monitor for real-time assessment of the sample.

Maximum sample resolution is obtained by placing the sample in close proximity to the x-ray aperture such that X-ray projection, a cone beam, narrowly spans the sample.
width. After the sample is scanned at one elevation, successive vertical translations and scanning of the sample are made until the desired sample length is obtained. The sample attenuation data is then converted to two-dimensional images (slices), which are later combined into single 3D images.

Figure 3.7  X-ray microtomography CT facility
(Courtesy from Navy Research Lab, Stennis Space Center, MS)

- Random Packing Experiments

The containers used for the packing experiments are an acrylic cylindrical container (23mm ID and 60mm depth) and a cubic container with rough inner surfaces ($L_{22} \text{mm}$, $W_{22} \text{mm}$, $H_{19} \text{mm}$). The PEEK/AL cylinders were placed into the containers. To obtain the lowest density packings that were analyzed in this study, the particles were added to the container while a vertical rod was in the center. Once the system was mounted on the XMT stage, the rod was removed slowly, allowing the particle to fall into a low-density but gravitational stable arrangement. A series of moderate-density packings were obtained simply by pouring the particles into the containers with varying degrees of gentleness. The highest density-packings were obtained by vibrating and tapping the sides of the container until no more settling was observed. A variety of vibration methods were attempted. It was found that more violent methods (e.g. a test-tube mixer) added too much energy to the system, bouncing the particles into low-density configurations. An
etching tool provided a gentler vibration, as did tapping on the acrylic containers with rigid metal rod once it was mounted firmly onto the XMT stage.

- Packing Reconstruction

Figure 3.8(a) is a grayscale image of a slice from the XMT imaging. The aluminum cores show brightly in the image, which allows them to be segmented from the PEEK tubing and air using simple thresholding of the grayscale image. The three-dimensional grayscale image is converted to a binary image by choosing a threshold x-ray absorbence between the value for the PEEK and the aluminum. Figure 3.8(b) depicts this three-dimensional binary image of the locations of the cylinder cores. Each core is composed of a series of cubic voxels.

To perform a detailed analysis of the packing structure, the XMT data are transformed into particle-scale data, i.e. the location and orientation of each cylinder in the packing. The first step in this particle reconstruction is a solid-phase burn of the data. A burn maps onto each voxel the (non-Euclidian) distance to the nearest surface of an aluminum core, which in turn can be used to identify individual cylinders within the voxelized data. The second step of the particle reconstruction involves a connected-
component analysis, which assembles clusters of voxels that are isolated from other clusters. If no Aluminum cores were touching (at least according to the resolution of the XMT data), a simple connected-component analysis would suffice to identify each single particle. Unfortunately, the data do show aluminum cores in contact, either because of true contacts in the packing or because of limited image resolution. Hence, some connected components identified in the first pass of the algorithm will represent more than one particle. Figure 3.9 is an example of a histogram of the number of connected components verses the number of voxels contained in each component. A strong peak is present near 8500, which is the predicted number of voxels per AL core for 0.0406mm resolution. Note however the existence of the secondary peaks at 2× and 3× the single-particle value. These peaks are indicative of larger clusters. An erosion is performing and the connected component algorithm is run once again (only on the large cluster of voxels). Voxels that were removed in the erosion process are added back onto the new connected component using a restricted burn procedure, and the size of these new clusters are checked to see whether they correspond to single particles. If any multiple-particle clusters remain, then this process is repeated using a level-two erosion. It continues until all particles are identified.

A couple of notes should be made. To run this process, one needs to bracket a range of connected-component sizes that are accepted as single particles. This range is chosen based on the predicted value (calculated from the known size of the Al cores and the known voxel resolution) combined with examination of the histogram for a given run. Second, there is no assurance that this procedure will work. For example, if two Al cores are in contact with their axes being collinear, the erosion is not capable of identifying the
two individual particles. However, in all cases tested to date, the procedure has converged. This result is due in large part to the high quality of the data, the high resolution in comparison to the cylinder size, and the apparent lack of any perfect end-to-end alignments in the packing tested.

![Histogram of the size of connected components](image)

**Figure 3.9** Histogram of the size of connected components

Once the set of individual AL cores are located, the position of each cylinder is recorded as the center of mass of its respective voxel cluster. The orientation of each cylinder is found by performing an optimization that minimizes the sum of the squares of the distance from the cylinder axis to each voxel in the cluster. In addition to the results shown here, the reconstruction algorithm was validated using computer-generated cylinder packings (locations and orientations known exactly from the original data set). The data were recovered to good accuracy for resolutions equivalent to the XMT data used in this work.

The particle-scale data are computed in Cartesian coordinates, and we keep the data in this form for convenience. The image data are listed in $x, y, z$ format, and all the
computations described above are independent of the container shape and size. The smoothness of the cylindrical domains (shown later) is a result of the accurate computation of particle positions and orientations.

3.2.2 Cylinder Packs in a Cylindrical Container

The other parameter that must be computed is the Cartesian location of the container axis (or $r=0$), which is necessary to plot parameters as a function of distance from the containing wall. The coordinates of the container axis are computed from the imaged data by an optimization algorithm. There are two steps of optimization process:

**Step 1:** Project all cylindrical particle centers into x-y plane. All of these scattered points are inside or on a circle, which is what we want to find. There must be at least three points on a circular perimeter whose diameter is smaller than the container inner diameter by a particle diameter ($D_{\text{particle}}$).

**Step 2:** There are two cases of the remaining problem: (a) If the container diameter is known in advance, a random search is performed to find the circle center that includes all the particle centers, and the global average of all the particle centers can be used as the starting point; (b) If the container diameter is unknown, the problem is to find a minimum circle that can hold all the particle centers (on or inside its perimeter). Both its center and radius are the targets. The key logic of the algorithm to perform this computation is randomly moving and shrinking a circle, while checking if it can hold all the centers. This process is continued until it cannot move or shrink. The initial circle is larger to have all points inside.

Five packs were imaged over what was believed to represent the widest possible range of packing density. Three packs were imaged initially at a lower voxel resolution of
0.0460mm. Two additional packs were added later at a higher voxel resolution of 0.0406mm. Also, more cylinders were added to assure the results are typical. The corresponding 3D computer-reconstructed images are shown in figure 3.10(a)-(e).

![Computer reconstructed five packs in a cylindrical container](image)

Figure 3.10  Computer reconstructed five packs in a cylindrical container

To compute the average density correctly, a top-crust (because of irregular top surface) and a bottom-crust (because of a regular face) have been cut off during computation. The choice of crust thickness is a two-particle-diameter distance each of top and bottom. In radial direction, the container radius size is chosen, in which the outmost layer represents the extreme near-wall property. Therefore, experimental packs need at least 5~6 times of $D_{\text{particle}}$ in height.

To study the wall effects, the packing is divided into two regions: an inner region and a near-wall region. This division is kind of arbitrary; the near-wall region is chosen as the wall distance of $\leq 2D_{\text{particle}}$; all other values are in the interior region. Thus, the
inner region better represents the bulk properties and the outer one represents the near-wall properties. (The domain is also divided into four regions from wall to the center, to study the density variations in different regions from the wall.)

The density computation is based on solid voxel fraction, and the whole domain is digitalized into 300×300×(250-350) small cubic voxels. This resolution is sufficient enough to match the computational cylinder volume with a relative small error less than 0.0002.

3.2.3 Cylinder Packs in a Cubic Container

We studied the cylinder packs in a cylindrical container to understand wall effects, while we studied the cylinder packs in a cubic container for bulk packing structure. Specifically, the rough walls are intentionally designed to eliminate any wall effects inside packs. Three packs were imaged at progressive densities during densification. All three packs have a voxel resolution of 0.0653mm, and the computer-reconstructed images of three rough-wall packs are shown in Figure 3.11(a)-(c).

![Figure 3.11 Computer reconstructed packs in a rough-wall container](image)

3.2.4 Preliminary Discussion

During densification, the particles undergo a series of settling and reorganizing processes. Their final configurations arise from a combination of gravity, friction, and
container geometry considerations. Detailed structural analysis based on particle scale information will involve particle positions, orientations, and their spatial correlations. Here, we provide some qualitative discussions, and a detailed structural analysis is reported in the next chapter.

- Density

While random sphere packings have been studied extensively, cylinder packings have not been systematically investigated. In quantifying the packing density, we will answer fundamental questions such as the values of the random loose and/or random close packing density, and whether experimental packs versus computer-simulated packings have equivalent structures. In addition, density variation in a cylindrical container is a relevant problem in chemical engineering (e.g. packed bed).

- Position

Positional ordering constitutes one aspect of structural characterizations. Simply projecting all cylinder centers onto the $x$-$y$ (or $x$-$z$ or $y$-$z$) plane provides a qualitative measurement of ordering. Projections of the experimental packs are shown in Figures 3.12 & 3.13.

![Projections of particle centers in a cubic container](image)

**Figure 3.12**  Projection of particle centers in a cubic container
Figure 3.13 Projection of particle centers in a cylindrical container

- Orientation

Orientational ordering constitutes another important aspect of structural properties. Qualitatively, we project all particle orientations onto a unit spherical surface, which gives a simple visualization of isotropic/anisotropic behavior. If all the points (representing particle orientation vectors) are uniformly scattered on the spherical
surface, we say the packing is isotropic; while if all the points are scattered in any symmetric manner, it is anisotropic. Spherical harmonics will be used to quantify this behavior later.

Figure 3.14  Projection of particle orientations in three rough-walled packs
CHAPTER 4
STRUCTURAL ANALYSIS OF RANDOM CYLINDER PACKINGS

The particle-scale packed structure influences fluid, heat, and mass transfer, which in turn affect macroscopic process and parameters such as reactor temperatures, flow maldistribution, separation efficiency, etc. For disordered packing of uniform-sized particles, the internal structure and porosity are dictated by the particle shape and arrangement of these particles during the packing process. The container geometry also influences the packing structure, e.g. the packing density near wall can differ significantly from the bulk density, which contributes to channeling problems in chemical engineering. Therefore, quantifying variations in particle-scale packing structure is important.

Several difficulties arise in characterizing the microscale structure for cylinder packings. Compared to spherical particles, cylinders exhibit more types of surface geometry (composed of angular, planar, and curved surfaces). Therefore, cylinder-cylinder contacts assume a variety of configurations. In addition, cylinders have rotational degrees of freedom.

In the last chapter of computer simulations, random dense packings were generated using the collective rearrangement method. The particles underwent a series of small random movements until the overlaps between them disappeared. The packing experiments are going to address the following questions: Can the computer-generated disordered packings duplicate the real experimental densification packs (or statistically equivalent structures)? During vibrating and tapping process, can particles rearrange
themselves to increase their local or global ordering? What is the possible phase state
equilibrium of disordered packing structures in this nonthermal particulate system?

In packing experiments, several interesting phenomena were observed. During
densification, slim cylinder pairs prefer to align themselves in parallel orientations
(Fernando, 2000; Montillet and Coq, 2001; Lumay and Vandewalle, 2004), while for
compact cylinders, perpendicular orientations were added (Zhang et al., 2005). The
physical boundary influences the orientations of cylinder packs, and the compact
cylinders near boundaries have preferred orientations (either end-contact or side-contact).
These interesting phenomena are natural combinations of gravity and frictional force.
However, it is very difficult to explain quantitatively some of basic questions, e.g. How is
the random dense packing density of equilateral cylinders (compared to sphere packings
usually used as a prototype model of granular materials)? How far can the orthogonal
inclination of compact cylinders extend influencing the global structures? How about the
physical boundary influences the inner structure? What are the structural (position and
orientation) changes as random packing undergo a series of densification processes? Can
compact cylinder packings generate strong pair correlation, which suggest layered
structure and long range of cubatic orders during densification? To answer above
questions, 3D detailed structural analyses are needed.

In this chapter, several types of disordered packing structures including both
computer simulated and experimental packings have been analyzed. In simulated
packings, all the detailed structural parameters of the RCPs were computed. In the
experimental packings, equilateral cylinders were used and a series of disordered packs in
two types of containers were performed. The cylindrical container is used to study the
wall effects and the rough-walled container is used to study the onset of ordering in the bulk structure. The specific objectives include: (a) compute all structural parameters of computer-simulated RCPs with varied aspect ratios; (b) compute all structural parameters of experimental packs in a cubic container during densification; (c) quantify the wall effects of experimental packs in a cylindrical container.

4.1 Structure of Simulated Packings

The computer-generated RCPs (see Table 3.1) are used in this section. All packings are believed to reach their packing limits, since the translational and rotational movements of particles were nearly constrained after collective rearrangement. Analysis of the final structure provides insight into the equilibrium state of non-thermal, nonspherical particulate systems. The detailed structural analysis includes density, positional ordering, local/global orientational ordering, and particle contact numbers.

4.1.1 Density

For computer-generated packings with periodic boundaries, the average density (or SVF) is a trivial computation. The ratio of the total volume of cylinders to the domain volume gives the average packing density. To obtain the varied density profiles (or how the packing density varies with the different locations), the cubic domain were discretized into a series of small shells and compute local densities. This process is based on small voxel computations. To be specific, the packing domain is discretized into a very large number of cubic-shaped voxels. Each voxel is assigned ‘1’ for solid or ‘0’ for void. Collecting all the voxels in a shell region and computing the ratio of solid voxel number to the total voxel number gives the local density. If a layered structure exists, there is a corresponding wave in the local density. (A series of cubic shells are used to discretize a
cubic domain that was used in simulated packings.) A schematic of the domain discretization in 2D is illustrated in Figure 4.1. The local density distributions versus wall distances are displayed in Figure 4.2.

Figure 4.1  A 2D schematic of domain discretization of a cube

Figure 4.2  Density distributions versus wall-distance in simulated packings  
\((L/D = 1, 5, 10 \text{ and } 20)\)
There is no density variation observed in near-wall regions in any of the figures. Part of the reason is that of the integrated periodicity in the simulated packings. It also suggests that the Monte Carlo perturbation process during collective rearrangement did not alter the homogeneous global structures. (The oscillation of the tail in figures 4.2 should be disregarded, because of the very small computational volume in the center of domain.)

4.1.2 Positional Ordering and RDF

The generalized RDF is used to quantify the positional ordering. During the computation, a pair separation is interpreted as a minimal distance from a cylinder center to the reference cylinder surface. (This definition is used instead of surface-surface distance to avoid the difficulty of calculating the average excluded volume of two cylinders, which will involves cylinder angle distributions and complicated integrals of the second kind (Blaak et al., 1999).) Therefore, an effective distance between two cylinders is from \(0.5D_{\text{particle}}\) to a finite large number (in a finite domain). Although one surface-surface distance can map several center-surface distances in random cylinder packings, this method is still effective in dealing with equilateral cylinders. Figure 4.3 lists the radial distribution functions of four simulated packings.

For \(L/D = 1\), the first strong peak occurs at \(0.5D_{\text{particle}}\) distance with an intensity value of \(~3.6\). The function also shows a distinct depletion region followed by a broad second peak with a value of \(~1.4\). The third peak is almost indiscernible. For \(L/D > 5\), the second peak almost disappears, which suggests that layered structures cannot extend farther. Note that we cannot conclude that even the first layer disappears for large aspect ratio packings, because larger particle \(L/Ds\) significantly dissipate the energy and scatter
in a wide pair distances. We can only conclude that local parallel (or nearly parallel) correlations are very weak. Note also that strong peaks in equilateral packings can be either in parallel or perpendicular configurations of cylinder pairs.

Figure 4.3  RDFs of simulated random packings ($L/D = 1, 5, 10, 20$)

4.1.3 Orientational Ordering

- Local Ordering

Local orientational ordering of computer-simulated RCPs were computed. The equilateral results will be used to compare with the experimental results. The formula used is based on 2\textsuperscript{nd}, 4\textsuperscript{th}, 6\textsuperscript{th}-order Legendre polynomials, and the independent variable is $\cos(\theta)$, which is the dot-product of two normalized vectors from two cylinder axes.
\[ S_2(\cos \theta) = \frac{1}{2}(3\cos^2 \theta - 1) \]

\[ S_4(\cos \theta) = \frac{1}{8}(35\cos^4 \theta - 30\cos^2 \theta + 3) \]

\[ S_6(\cos \theta) = \frac{1}{16}(231\cos^6 \theta - 315\cos^4 \theta + 105\cos^2 \theta - 5) \]

The neighboring pair is defined where the minimal distance between two cylinders is less than its diameter \(D_{\text{particle}}\) (i.e., no another particle can be inserted between them). The corresponding results are listed in Table 4.1. In addition, local ordering was also computed when the pair distance is set a smaller value of \(0.05D_{\text{particle}}\) (see Table 4.2). Physically, we expect neighboring cylinders to surround the base cylinder on its geometric contour. The derived results help to explain what kind of equilibrium state will be reached through Monte Carlo perturbation during collective rearrangement in cylindrical particulate systems.

**Table 4.1 Local orders (when \(gap = D_{\text{particle}}\))**

<table>
<thead>
<tr>
<th>Packing</th>
<th>Local (S_2)</th>
<th>Local (S_4)</th>
<th>Local (S_6)</th>
<th># Neighbors</th>
<th>(L/D)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2_5_1*</td>
<td>1.191E-3</td>
<td>0.0642</td>
<td>7.527e-4</td>
<td>37.2</td>
<td>1</td>
</tr>
<tr>
<td>2_10_1</td>
<td>-0.006588</td>
<td>0.04975</td>
<td>0.00473</td>
<td>37.5</td>
<td>1</td>
</tr>
<tr>
<td>H11_65</td>
<td>0.2733e-4</td>
<td>0.0362</td>
<td>-0.571e-3</td>
<td>35.5</td>
<td>1.1</td>
</tr>
<tr>
<td>H12_66</td>
<td>-0.005428</td>
<td>0.0342</td>
<td>0.00186</td>
<td>34.9</td>
<td>1.2</td>
</tr>
<tr>
<td>H15_645</td>
<td>-0.4008e-3</td>
<td>0.00542</td>
<td>0.00941</td>
<td>31.7</td>
<td>1.5</td>
</tr>
<tr>
<td>2_5_2</td>
<td>-0.00895</td>
<td>0.00875</td>
<td>-0.00502</td>
<td>28.5</td>
<td>2</td>
</tr>
<tr>
<td>2_2_5</td>
<td>-0.004944</td>
<td>0.01164</td>
<td>0.005346</td>
<td>24.2</td>
<td>5</td>
</tr>
<tr>
<td>2_4_5</td>
<td>-0.03609</td>
<td>0.009082</td>
<td>0.006302</td>
<td>25.5</td>
<td>5</td>
</tr>
<tr>
<td>2_2_8</td>
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<td>0.01097</td>
<td>-0.002897</td>
<td>22.4</td>
<td>8</td>
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<tr>
<td>2_2_10</td>
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<td>0.008576</td>
<td>0.01004</td>
<td>20.7</td>
<td>10</td>
</tr>
<tr>
<td>2_2_16</td>
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<td>0.01294</td>
<td>0.007994</td>
<td>19.7</td>
<td>16</td>
</tr>
<tr>
<td>2_2_20</td>
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<td>0.005126</td>
<td>0.004755</td>
<td>17.7</td>
<td>20</td>
</tr>
<tr>
<td>2_1_50</td>
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<td>0.04146</td>
<td>-0.002317</td>
<td>14.6</td>
<td>50</td>
</tr>
<tr>
<td>2_2_50</td>
<td>-0.1745</td>
<td>0.004943</td>
<td>0.008211</td>
<td>16.7</td>
<td>50</td>
</tr>
<tr>
<td>2_1_100</td>
<td>-0.2182</td>
<td>0.03730</td>
<td>-0.01169</td>
<td>14.1</td>
<td>100</td>
</tr>
</tbody>
</table>

Note: 2_5_1* represents the average from five equilateral packings.
Table 4.2  Local orders (when $gap=0.05D_{\text{particle}}$)

<table>
<thead>
<tr>
<th>Packing</th>
<th>Local $S_2$</th>
<th>Local $S_4$</th>
<th>Local $S_6$</th>
<th># Neighbors</th>
<th>$L/D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2_5_1*</td>
<td>-1.919e-2</td>
<td>0.1699</td>
<td>0.05636</td>
<td>7.6</td>
<td>1</td>
</tr>
<tr>
<td>2_10_1</td>
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<td>0.1578</td>
<td>0.004678</td>
<td>8.1</td>
<td>1</td>
</tr>
<tr>
<td>H11_65</td>
<td>-0.05642</td>
<td>0.1472</td>
<td>-9.406e-3</td>
<td>7.5</td>
<td>1.1</td>
</tr>
<tr>
<td>H12_66</td>
<td>-9.205e-3</td>
<td>0.109</td>
<td>-5.202e-3</td>
<td>7.0</td>
<td>1.2</td>
</tr>
<tr>
<td>H15_645</td>
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<td>0.05906</td>
<td>0.02521</td>
<td>5.9</td>
<td>1.5</td>
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<tr>
<td>2_5_2</td>
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<td>0.04439</td>
<td>-0.01172</td>
<td>6.7</td>
<td>2</td>
</tr>
<tr>
<td>2_2_5</td>
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<td>0.07048</td>
<td>0.04635</td>
<td>5.88</td>
<td>5</td>
</tr>
<tr>
<td>2_4_5</td>
<td>-5.527e-3</td>
<td>0.03357</td>
<td>0.03376</td>
<td>6.9</td>
<td>5</td>
</tr>
<tr>
<td>2_2_8</td>
<td>-0.1166</td>
<td>0.0615</td>
<td>0.0144</td>
<td>5.5</td>
<td>8</td>
</tr>
<tr>
<td>2_2_10</td>
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<td>0.05047</td>
<td>0.02264</td>
<td>4.3</td>
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</tr>
<tr>
<td>2_2_16</td>
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<tr>
<td>2_2_20</td>
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<td>0.02326</td>
<td>9.164e-3</td>
<td>3.4</td>
<td>20</td>
</tr>
<tr>
<td>2_1_50</td>
<td>-0.1994</td>
<td>0.04156</td>
<td>-0.01694</td>
<td>2.6</td>
<td>50</td>
</tr>
<tr>
<td>2_2_50</td>
<td>-0.1729</td>
<td>7.2103e-3</td>
<td>0.01216</td>
<td>2.3</td>
<td>50</td>
</tr>
<tr>
<td>2_1_100</td>
<td>-0.223</td>
<td>0.0317</td>
<td>-6.395e-3</td>
<td>2.5</td>
<td>100</td>
</tr>
</tbody>
</table>

Note: 2_5_1* represents the average from five equilateral packings.

The results from above tables can be summarized as follows--

a. The gap is the surface-surface distance between particles. Larger gap means more particles are included to form a bigger particle cluster (as viewed).

b. Local $S_2$ represents a parallel correlation, which does not show up in compact (e.g. $L/D \leq 2$) cylinder packings. For larger aspect ratios (e.g. $L/D >10$), the packing exhibits certain local correlations, which are negative suggesting a denser pair distribution for larger angles (>55°) but sparser for small angles (<55°).

Further inferences cannot be made.

c. Local $S_4$ represents orthogonal correlations, which only exist in the packings of compact cylinders (see Table 4.2). The correlation almost disappears for aspect ratios larger than 1.5, suggesting the phenomenon is unique for the equilateral geometry. As the pair-distance increases, this correlation disappears (see Table
4.1). A safe conclusion is that orthogonal correlations are very weak for simulated packing, though Blaak et al. (1999) obtained in their simulation by introducing a flip-move.

d. No local $S_6$ ordering has observed since all the values are very small.

e. The average number of neighbors decreases steadily with particle aspect ratio. This result is inconsistent with theory (Philipse, 1986), which predicts a flat region of the contact number as the aspect ratio becomes larger than 15. This is one of the disadvantages of using the collective rearrangement method, since RCP does not generate physically stable structures. Near the equilateral region, more and more flat-end surface involves in the pair minimal distance. To better understand the contact number in simulated random close packings, a new analysis is given in a later section.

- **Global Ordering**

  Global ordering is computed using spherical harmonics and Legend Polynomials. Spherical harmonics have special properties allowing them to evaluate complicated 3d symmetric structures. Simple algebraic manipulations provide scalar results with good physical interpretations. Specifically, the quadratic invariant parameters $I_2$ and $I_4$ were computed to examine possible nematic and cubatic information.

  The quadratic invariant combination of spherical harmonics is used to evaluate symmetric information in the packing, which has the ability to express uniaxial, biaxial and cubatic symmetric structures. It is defined

  $$Y^m_l(\theta, \phi) = \frac{2l + 1}{4\pi} \frac{(l - m)!}{(l + m)!} P^m_l(\cos \theta) e^{im\phi}$$

  ----spheric harmonic
where $\theta$ is the zenith, $\phi$ is the azimuth and $P_i^m$ are the associated Legendre polynomials.

Two sets of other formulas were also used to quantify the global ordering: one is the simple pair correlation (Coelho, 1997) and the other is from liquid crystal theory: $2^{nd}$, $4^{th}$-order Legend polynomials. The results are listed in Table 4.3.

<table>
<thead>
<tr>
<th>Packing</th>
<th>$M$</th>
<th>$S_2$</th>
<th>$S_4$</th>
<th>$I_2$</th>
<th>$I_4$</th>
<th>$I_6$</th>
<th># Particles</th>
<th>$L/D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2_5_1*</td>
<td>0.0001</td>
<td>0.0815</td>
<td>0.0362</td>
<td>0.0952</td>
<td>0.2236</td>
<td>0.0970</td>
<td>103</td>
<td>1</td>
</tr>
<tr>
<td>2_10_1</td>
<td>-3.957e-4</td>
<td>0.0259</td>
<td>0.0118</td>
<td>0.0285</td>
<td>0.0647</td>
<td>0.0377</td>
<td>827</td>
<td>1</td>
</tr>
<tr>
<td>H11_65</td>
<td>-8.938e-4</td>
<td>0.0624</td>
<td>-0.0629</td>
<td>0.0843</td>
<td>0.1362</td>
<td>0.1074</td>
<td>125</td>
<td>1.1</td>
</tr>
<tr>
<td>H12_66</td>
<td>-7.769e-4</td>
<td>0.0672</td>
<td>0.0437</td>
<td>0.0765</td>
<td>0.1233</td>
<td>0.0815</td>
<td>151</td>
<td>1.2</td>
</tr>
<tr>
<td>H15_645</td>
<td>-6.789e-4</td>
<td>0.0562</td>
<td>-0.0259</td>
<td>0.0606</td>
<td>0.0974</td>
<td>0.0863</td>
<td>230</td>
<td>1.5</td>
</tr>
<tr>
<td>2_5_2</td>
<td>-0.0018</td>
<td>0.0236</td>
<td>-0.0158</td>
<td>0.0286</td>
<td>0.0372</td>
<td>0.0382</td>
<td>381</td>
<td>2</td>
</tr>
<tr>
<td>2_2_5</td>
<td>0.0042</td>
<td>0.1023</td>
<td>-0.0649</td>
<td>0.1083</td>
<td>0.1375</td>
<td>0.095</td>
<td>132</td>
<td>5</td>
</tr>
<tr>
<td>2_4_5</td>
<td>-1.498e-4</td>
<td>0.0255</td>
<td>0.0414</td>
<td>0.0285</td>
<td>0.0744</td>
<td>0.0283</td>
<td>1038</td>
<td>5</td>
</tr>
<tr>
<td>2_2_8</td>
<td>0.0012</td>
<td>0.0473</td>
<td>0.0222</td>
<td>0.0697</td>
<td>0.0783</td>
<td>0.0531</td>
<td>273</td>
<td>8</td>
</tr>
<tr>
<td>2_2_10</td>
<td>-0.0013</td>
<td>0.029</td>
<td>0.0179</td>
<td>0.0386</td>
<td>0.0829</td>
<td>0.0775</td>
<td>356</td>
<td>10</td>
</tr>
<tr>
<td>2_2_16</td>
<td>-7.884e-4</td>
<td>0.0249</td>
<td>0.0725</td>
<td>0.0263</td>
<td>0.1072</td>
<td>0.0388</td>
<td>677</td>
<td>16</td>
</tr>
<tr>
<td>2_2_20</td>
<td>-3.491e-4</td>
<td>0.0222</td>
<td>0.0102</td>
<td>0.0297</td>
<td>0.0859</td>
<td>0.0299</td>
<td>814</td>
<td>20</td>
</tr>
<tr>
<td>2_1_50</td>
<td>-0.0015</td>
<td>0.033</td>
<td>0.0261</td>
<td>0.0363</td>
<td>0.1201</td>
<td>0.0593</td>
<td>350</td>
<td>50</td>
</tr>
<tr>
<td>2_2_50</td>
<td>9.141e-5</td>
<td>0.0159</td>
<td>0.0468</td>
<td>0.0220</td>
<td>0.102</td>
<td>0.0264</td>
<td>2546</td>
<td>50</td>
</tr>
<tr>
<td>2_1_100</td>
<td>-7.71e-4</td>
<td>0.0236</td>
<td>-0.0129</td>
<td>0.0257</td>
<td>0.1023</td>
<td>0.0241</td>
<td>700</td>
<td>100</td>
</tr>
</tbody>
</table>

Note: 2_5_1* represents the average from five equilateral packings.

No conspicuous global ordering is evident, since all the values are smaller than 0.3 (which is believed to be the lower transitional boundary in liquid crystal theory).

Therefore, collective rearrangement does NOT obviously modify the global structures of disordered packings. The reason is likely the logic in the CR algorithm, e.g. the random perturbation was limited in a small spherical region regulated by the cylinder diameter. A
small degree of orthogonal symmetry appears to exist in some compact cylinder packings (e.g. 2-5-1*), and further explain the relevant results are difficult.

4.1.4 Particle Contact Number

Average contact number was mentioned relative to local ordering computations. Now it is studied in detail. Contact number of a cylinder in a random pack is important in predicting the random packing density theoretically, because it reflects the physical contacts necessary to cage a cylinder (Williams & Philipse, 2003). Computer-simulated packings are not in physical contacts, but with a small numerical error, qualitatively predict the contact number and its trend in the packings is possible. The contact number versus minimal distance in different packings is displayed in Figure 4.4.

![Figure 4.4 Contact numbers versus minimal distance (L/D=1,5,10,20)](image)

Figure 4.4 Contact numbers versus minimal distance (L/D=1,5,10,20)
Philipse (2003) proposed that, for a given particle shape, the packing density is determined by the average number of contacts. This relationship was expressed in a random contact equation

$$\phi E = \langle \gamma \rangle = \frac{1}{N} \sum_{i}^{N} \gamma_i$$

where $\phi$ is the packing density, $E$ is a fixed number and $\gamma$ is the contact number. The average minimal number of neighbors to immobilize a particle by caging in random sphere packings is $\langle \gamma \rangle = 4.79$, while to achieve such caging in spherocylinder packing, $\phi \alpha = \langle \gamma \rangle \geq 5$ is required. Our simulation results are lower than the theoretical prediction.

4.2 Structure of Experimental Packs in a Rough-Walled Container

In this section, equilateral cylinder packs in a cubic container with rough surfaces have been examined. The rough surfaces are designed intentionally to avoid any wall effects, and the results are used to compare with the computer-simulated results. Specific objectives include: (a) Find the practical bulk dense packing density of equilateral cylinders; (b) Explore any positional ordering (locally or globally); (c) Quantify any orientational ordering and its evolution during densification.

Three rough-wall packs were imaged and the reconstructed images were shown in Figure 3.11. These three packs correspond to an increasing packing density. For convenience, they are named R1, R2 and R3 respectively in the following discussions.

4.2.1 Radial Density Profile

The packings obtained from XMT are approximately in a shape of long box (see Figure 3.11). The computational domain is a smaller box with a square cross-section in the $x$-$y$ plane, which is derived by cutting $1.5D_{\text{particle}}$ layers from each side and $2D_{\text{particle}}$ layers
from the top and bottom. This choice removes rough layer surfaces in packings. The computational process is based on the solid voxel fraction (same procedure as used before). The density distributions in radial and $z$ directions are illustrated in Figure 4.5 below.

Figure 4.5  Density distributions in rough-walled packings
(Left: radial density in R1, R2, R3 packings. Right: $z$ density in R1, R2, R3 packings)
The above figures suggest that the wall effects were effectively removed in the rough-walled packs, since the density is nearly constant everywhere. The average densities for three packings are 0.644 (R1), 0.663 (R2) and 0.700 (R3).

4.2.2 Positional Ordering

Not all the particles can be used as the reference particles because the real structures are not periodic. In the computation, 20 cylinders in the middle region were chosen to calculate the particle RDFs. The derived plots are illustrated in Figure 4.6.

![Particle RDF plots](image)

**Figure 4.6** Particle RDFs of three rough-walled packings (R1, R2, R3)

The particle RDFs suggest that the structure in equilateral cylinder packs extends about three layers at most. The first sharp peak occurs at \(0.5D_{\text{particle}}\) and then wider small
peaks follow at $1.5D_{\text{particle}}$ and $2.5D_{\text{particle}}$. After three layers, it degenerates to 1. At least two peaks are obvious, suggesting that the real packing cannot be completely random. A small third peak appears in R2 pack and a depletion region (RDF<1) appears following the first peak. The first peak gains intensity (4.4→4.7), as the pack becomes denser during densification, suggesting particle natural inclination. All three intensities are larger than that of simulated packings. The depletion region is seen clearly in R3 pack but it is not as wide as sphere packing.

4.2.3 Orientational Ordering

The same formulas described previously are use to calculate the orientational ordering in the three packings discussed here. For local ordering, two gaps $2D_{\text{particle}}$ and $0.05D_{\text{particle}}$ are used; for global ordering, a new set of correlations $Q_2$ and $Q_4$ (parallel and orthogonal with the gravitational vector) are added.

- **Local Orders**

| Table 4.4  Local ordering of three rough-walled packs (gap=$D_{\text{particle}}$) |
|-----------------|----------------|----------------|----------------|----------------|----------------|
| Packing        | Local $S_2$   | Local $S_4$   | Local $S_6$   | # Neighbors    | L/D            |
| R1             | 6.321e-3      | 6.509e-2      | 4.838e-2      | 25.5           | 1.04           |
| R2             | 8.524e-3      | 8.441e-2      | 5.328e-2      | 26.0           | 1.04           |
| R3             | 8.656e-3      | 0.1072        | 0.1143        | 28.3           | 1.04           |

| Table 4.5  Local ordering of three rough-walled packs (gap=0.05$D_{\text{particle}}$) |
|-----------------|----------------|----------------|----------------|----------------|----------------|
| Packing        | Local $S_2$   | Local $S_4$   | Local $S_6$   | # Neighbors    | L/D            |
| R1             | 1.245e-2      | 0.2267         | 9.40e-2       | 4.1            | 1.04           |
| R2             | -9.013e-3     | 0.2555         | 0.1042        | 4.3            | 1.04           |
| R3             | 7.949e-3      | 0.2747         | 0.1668        | 5.0            | 1.04           |

From tables 4.4 & 4.5, it is observed that small particle groups (5-6 cylinders) have a certain degree of local ordering (gap=0.05$D_{\text{particle}}$) in all packings; when these groups are extended farther (26-30 cylinders), most local ordering disappears. During
densification, local $S_4$ and $S_6$ ordering increases and the number of particle neighbors goes up.

Comparing local ordering results from experimental packs (Tables 4.4 & 4.5) with the results from the computer-simulated packings (Tables 4.1 & 4.2), it is found that experimental packs exhibit moderate local ordering (e.g. $S_4$ and $S_6$ in R3), whereas simulated packings do not exhibit ordering. Surprisingly, experimental packings have fewer numbers of neighbors. These phenomena suggest that the Monte Carlo perturbations during collective rearrangement cannot reproduce the complexity of real structures.

- **Global Ordering**

The global ordering in three packings is displayed in Table 4.6 below.

<table>
<thead>
<tr>
<th>Packing</th>
<th>$M$</th>
<th>$Q_2$</th>
<th>$Q_4$</th>
<th>$S_2$</th>
<th>$S_4$</th>
<th>$I_2$</th>
<th>$I_4$</th>
<th>$I_6$</th>
<th># Particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>R1</td>
<td>-7.14e-4</td>
<td>-0.007</td>
<td>-0.126</td>
<td>0.0239</td>
<td>0.0648</td>
<td>0.0316</td>
<td>0.134</td>
<td>0.0449</td>
<td>585</td>
</tr>
<tr>
<td>R2</td>
<td>-1.20e-3</td>
<td>-0.008</td>
<td>-0.122</td>
<td>0.0152</td>
<td>-0.026</td>
<td>0.0213</td>
<td>0.137</td>
<td>0.0376</td>
<td>612</td>
</tr>
<tr>
<td>R3</td>
<td>3.82e-4</td>
<td>-0.002</td>
<td>-0.122</td>
<td>0.042</td>
<td>0.0732</td>
<td>0.043</td>
<td>0.139</td>
<td>0.0457</td>
<td>681</td>
</tr>
</tbody>
</table>

From Table 4.6, there is no evidence of pair correlation ($M$), gravitational effects ($Q_2$ & $Q_4$), long-range nematic, or orthogonal ordering. Hence, the rough-walled packings can be classified as disordered (or random) in general sense.

4.2.4 Particle Contact Number

The inaccuracy of contact numbers comes from the resolution of X-ray tomography, cylinder shape in the fabrication, numerical errors during model reconstruction, and the calculation. Using current resolution data (65.3µm per voxel), we
can estimate the particle contact numbers in the packing. The error caused from voxel size is 3.63% of the cylinder diameter (1800µm), so the real contacts are shifted to the values corresponding to ~3.6% of minimal distance. From Figure 4.7, the contact numbers in three packs are 4.6 (R1), 4.8 (R2) and 5.3 (R3).

Comparing the contact numbers for experimental packings (e.g. R3 in Figure 4.7) with compute-simulated packings (e.g. \( L/D =1 \) in Figure 4.4), the latter has a steeper start. The contact number is 3.7–7.4 for 0-3.6%\( D_{\text{particle}} \) of minimal distance. These numbers are larger than the numbers from the R3 packing (3.4–5.3), which suggests that the simulated equilateral packings have different structural information in contact numbers.

### 4.3 Structure of Experimental Packs in a Cylindrical Container

In this section, equilateral cylinder packs in a cylindrical container with smooth surfaces have been examined. The specific objectives include: (a) Compute the density variation in the radial direction in a fixed bed; (b) Explore any positional ordering where there is wall effects during densification experiments; (c) Quantify any orientational ordering during densification.

The detailed computational parameters used to quantify the structure include global ordering (\( I_2 \) and \( I_4 \)), local ordering (\( S_2 \) or \( S_4 \)), wall RDF, particle RDF, and radial
density function. The computational procedure is same as above, so it is skipped.

However, new formulas must be designed to show correlations in a cylindrical container (wall effects) and the radial distribution functions. The formulas should reflect the possible alignment of particles with \( r, \phi, \) and \( z \) three directions. Using Legendre polynomials, they are defined

\[
W_{4,r} = \frac{1}{8} \left\{ 35 \cos^4 \theta_{i,r} - 30 \cos^2 \theta_{i,r} + 3 \right\}, \quad \text{scale 0–1}
\]

\[
W_{4,\phi} = \frac{1}{8} \left\{ 35 \cos^4 \theta_{i,\phi} - 30 \cos^2 \theta_{i,\phi} + 3 \right\}, \quad \text{scale 0–1}
\]

\[
W_{4,z} = \frac{1}{8} \left\{ 35 \cos^4 \theta_{i,z} - 30 \cos^2 \theta_{i,z} + 3 \right\}, \quad \text{scale 0–1}
\]

4.3.1 Radial Density

The radial density variation versus wall distance is based on the voxel computations. Because of the cylindrical container shape, circular rings were chosen for the computation of radial (local) density. To obtain the center axis of a pack, a numerical algorithm is applied (see 3.2.2). Because of the unpacked surface and flat bottom, a cropped working domain in \( z \) direction is used to ensure reliable results. In practice, two cylinder diameter distances were cut off to remove the end effects.

The radial density functions versus wall-distance are displayed in Figure 4.8. The five packs (C1, C2, C3, C4 & C5) are in a sequence of increasing density. A new non-dimensional wall distance \( (\xi = R_{\text{container}} - r)/D_{\text{particle}} \) is defined in units of the averaged cylinder diameter, which will be used frequently in the remaining analysis.
Figure 4.8  Radial density function versus wall-distance $\xi (C_1, C_2, C_3, C_4, C_5)$

From Figure 4.8, it is found that the wall effects can create correlated or periodic variations in density. For the low-density packing, the wall effects reach approximately 2-3 particle diameter distance into the interior of the packing. For the high-density packing,
the wall effects are present at least five particle diameters into the center. As the packing density increases, the functionality in the density versus wall-distance curve becomes steeper, which is indicative of the ability for the cylinder particles to line up nearly flat against the container wall (end-wall contact or side-wall contact). In contrast, the lower density packings exhibit a greater number of corner-wall contacts.

To study the detailed packing density in discrete regions, the wall distance was divided into four equal regions in the radial direction. Thus, different packing densities in different regions represent corresponding packing densities with differing wall effects. The regions are labeled in ascending order starting from the wall; the largest one represents the center density with the smallest wall effects. The packing densities are given in Table 4.7.

### Table 4.7 Packing densities in different regions

<table>
<thead>
<tr>
<th>Packing</th>
<th>Region 1</th>
<th>Region 2</th>
<th>Region 3</th>
<th>Region 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>0.5290</td>
<td>0.6569</td>
<td>0.6348</td>
<td>0.6145</td>
</tr>
<tr>
<td>C2</td>
<td>0.5802</td>
<td>0.6718</td>
<td>0.6691</td>
<td>0.6624</td>
</tr>
<tr>
<td>C3</td>
<td>0.6074</td>
<td>0.6927</td>
<td>0.6962</td>
<td>0.6952</td>
</tr>
<tr>
<td>C4</td>
<td>0.6762</td>
<td>0.7228</td>
<td>0.6846</td>
<td>0.7295</td>
</tr>
<tr>
<td>C5</td>
<td>0.6901</td>
<td>0.7348</td>
<td>0.7333</td>
<td>0.7358</td>
</tr>
</tbody>
</table>

### Table 4.8 Packing densities in interior and near-wall regions

<table>
<thead>
<tr>
<th>Packing</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Inner region</td>
</tr>
<tr>
<td>C1</td>
<td>0.6354</td>
</tr>
<tr>
<td>C2</td>
<td>0.660</td>
</tr>
<tr>
<td>C3</td>
<td>0.685</td>
</tr>
<tr>
<td>C4</td>
<td>0.6932</td>
</tr>
<tr>
<td>C5</td>
<td>0.7208</td>
</tr>
</tbody>
</table>
The packing is also divided into two regions representing near-wall and interior locations. The division surface is at $\xi = 2$. The results are displayed in Table 4.8. A significant observation from the quantitative results is that the packing density in the near-wall region increases faster than the bulk region during densification. For the low-density packing, the near-wall density is significantly lower than the bulk density. However, as the packing become denser, the near-wall density is closer to the bulk density. The reason for this evolution is the local cylinder rearrangement near the physical wall: cylinders either choose side-contact or head-contact with the wall and generate ordered structures (see Figure 4.9).

![Figure 4.9 Illustration of highly ordered structure in cylinder packings](image)

It is possible that a reversal of the local density could happen as the overall packing density continues to increase. Specifically, for an ideal regular structure of orthogonal configuration, the packing density is equal everywhere, which is higher than the bulk internal density. However, this perfect structure has not been observed in real packs because of many irregular-positioned particles present in the packing structures.
4.3.2 RDF and Positional Ordering

The wall RDF versus wall-distance ($\zeta$) plots are shown in Figure 4.10, which quantitatively express the layered distribution of particle centers shown in Figure 3.12.

Figure 4.10  Wall RDFs versus $\zeta$ in five packs (C1,C2,C3,C4,C5)

The particle RDF provides information about changes in local structure that evolve during densification. However, this parameter is of more interest in the context of
bulk packing structure (away from the walls). Because of the relative number of cylinders involved in the experiments, 26 cylinders in the middle were chosen for the computation. The derived RDF versus pair-distance plots are displayed in Figure 4.11.

Figure 4.11  Particle RDFs versus $\zeta$ in five packs (C1,C2,C3,C4,C5)
Figures 4.10 & 4.11 suggest that the positional ordering is a combined effect from the container walls and from neighboring particles. The container walls have a stronger influence (extending 3~5 layers) while neighboring particles have a weak influence (obvious at the first layer but quickly decreasing in the second to third layers). As the packing density increases, both of the influences can reach deeper and farther, and the packing becomes more regular.

4.3.3 Orientational Ordering

• Radial Ordering

Experimental observation tells that equilateral cylinders in the near-wall region have very strong orthogonal correlation with the $r$ direction. $S_{4,r}$ can also be computed as a function of wall distance ($\xi$). The correlation function is the 4$^{th}$-order Legendre polynomial, aligned with the $r$ direction. The radial ordering results for five packs are displayed in Figure 4.12. The results are also shown as a cumulative $S_{4,r}$ function, as the computational domain changed slowly from a small ring region near the wall to the whole packing (Figure 4.13). This helps quantify the near-wall and global radial ordering.

Figure 4.12  Radial ordering versus wall distance in five packs (C1,C2,C3,C4,C5)
Combing the radial ordering and the structural information from the RDFs shows that cylinders prefer to line themselves up in layered positional ordering and also prefer a strong orthogonal orientational ordering in $r$ direction. The decreasing peaks along $\xi$ imply that the radial ordering structure weakens as the ring (layer) moves from the wall toward the center (Figure 4.12). As the packings density increases, the maximum ordering increases, explaining the easier rearrangement of cylinders near the wall than the ones in the bulk center. Between the peaks, negative values are observed, which probably suggests that a very small number of cylinders were squeezed in anti-radial-correlated orientations between layers.
The cumulative radial ordering shown in Figures 4.13 suggests that both local ordering near the wall and global ordering steadily increase during densification, and the near-wall ordering is much larger than the bulk ordering.
- V-Ordering

Visual inspection suggests that a V-like structure exists in the experimental packs. That is, the particles oriented orthogonal to \( r \) and at roughly 45° with the horizontal plane. To verify this structure, a new ordering parameter \( S_{4,\alpha} \) is defined and calculated. Specifically, a vector on the \( \theta-z \) plane is defined and it has an angle with the horizontal. This vector is then correlated with particle orientations using the fourth-order Legendre polynomial. The results of V-ordering parameters \( S_{4,\alpha} \) versus \( \alpha \) in three packs are shown in Figure 4.14.

![Figure 4.14 V-ordering versus \( \alpha \) in three packs (C1, C3, C5)](image)

Figure 4.14  V-ordering versus \( \alpha \) in three packs (C1, C3, C5)
The distinct maxima (at $\alpha=45^\circ$) in three packs show the existence of V-ordering. The physical origin of this phenomenon is the favor of the ring-like structures, in which the radial staggering of particles is minimized and the maximum numbers of particle centers are located at the wall-distance $\zeta=0.5, 1.5, 2.5$ etc. (Zhang, et al., 2006).

The cumulative V-ordering function versus $\zeta$ in five experimental packs is shown in Figure 4.15.

![Figure 4.15 Cumulative V-ordering of five packs (C1,C2,C3,C4,C5)](image)

- **Local Ordering**

  Following the same computational procedure, local orientational ordering was systematically computed for five experimental packs. Due to wall effects, the results reflect the combined influence of particle-particle rearrangement and particle-wall rearrangement. The local ordering results are displayed in Tables 4.9 & 4.10.

  The local ordering in Tables 4.9 & 4.10 gives very interesting results: the local orientational ordering in five experimental packs is larger than the local ordering in simulated packings of equilateral cylinders. The local orthogonal correlation ($S_4$) in the
Experimental packs is very conspicuous, e.g. the ordering parameter in C5 pack is larger than 0.4. Another phenomenon noticed is that a local icosahedral correlation ($S_6$) emerges when the packing becomes dense, while the computer simulations do not show this specific phenomenon. The values of average number of particle neighbors are problematic since the experiment is non-periodic and many cylinders near the boundary (wall, top or bottom) are included. We will address the particle contact number in detail later.

<table>
<thead>
<tr>
<th>Packing</th>
<th>Local $S_2$</th>
<th>Local $S_4$</th>
<th>Local $S_6$</th>
<th># Neighbors</th>
<th>$L/D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>-0.001</td>
<td>0.056</td>
<td>0.038</td>
<td>27.4</td>
<td>1.079</td>
</tr>
<tr>
<td>C2</td>
<td>0.001</td>
<td>0.107</td>
<td>0.054</td>
<td>26.7</td>
<td>1.04</td>
</tr>
<tr>
<td>C3</td>
<td>-0.001</td>
<td>0.121</td>
<td>0.047</td>
<td>28.1</td>
<td>1.04</td>
</tr>
<tr>
<td>C4</td>
<td>0.002</td>
<td>0.167</td>
<td>0.046</td>
<td>28.7</td>
<td>1.04</td>
</tr>
<tr>
<td>C5</td>
<td>-0.001</td>
<td>0.184</td>
<td>0.103</td>
<td>32.4</td>
<td>1.04</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Packing</th>
<th>Local $S_2$</th>
<th>Local $S_4$</th>
<th>Local $S_6$</th>
<th># Neighbors</th>
<th>$L/D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>-0.012</td>
<td>0.193</td>
<td>0.073</td>
<td>4.5</td>
<td>1.079</td>
</tr>
<tr>
<td>C2</td>
<td>-0.008</td>
<td>0.286</td>
<td>0.136</td>
<td>4.4</td>
<td>1.04</td>
</tr>
<tr>
<td>C3</td>
<td>0.013</td>
<td>0.302</td>
<td>0.115</td>
<td>4.8</td>
<td>1.04</td>
</tr>
<tr>
<td>C4</td>
<td>-0.012</td>
<td>0.331</td>
<td>0.095</td>
<td>4.9</td>
<td>1.04</td>
</tr>
<tr>
<td>C5</td>
<td>0.003</td>
<td>0.383</td>
<td>0.224</td>
<td>6.0</td>
<td>1.04</td>
</tr>
</tbody>
</table>

To study further how local ordering varies within rings, local nematic and cubatic ordering functions versus $\xi$ (in five experimental packs) were computed. A particle is included in the computation as long as the particle center is inside of a ring. The results are shown in Figure 4.16. The nematic ordering parameters show no significant preferential alignment in five packs, which imply that there is not a strong tendency to align side-by-side or end-to-end locally. The local orthogonal ordering is finite.
throughout the interior region in all five packs, which indicates the tendency of the equilateral cylinders to lie at orthogonal orientations relative to one another. For the high-density packings, the orthogonal ordering parameter becomes oscillatory. This behavior is indicative of the strong orthogonal ordering near the wall that is evident in the figures. Equally interesting is that the value drops to zero between the peaks. This behavior suggests that in the relatively rare cases that a particle lies between one of the concentric rings (see the RDF plots), these particular particles are unlikely to be orthogonal to the neighboring structure. The inference is that they are more likely to lie at an odd angle when they are squeezed into a non-lattice position within the layer structure.

Figure 4.16  Local nematic and orthogonal ordering in five packs
(Left: local nematic ordering;  Right: local orthogonal ordering)

(fig con’d)
Global Ordering

Global orientational ordering was computed using spheric harmonics, which is based on the Cartesian coordinate system. The experimental results here include the combined influence from the wall effects. The circular wall forced the near-wall particles to rearrange their orientations, and it is possible to have certain symmetric structures in
the cylindrical system while it is not present in Cartesian system. For completeness, global orientational ordering was computed. The results are displayed in Table 4.11. The results show very weak global orientational ordering in the experimental packs (only C4 pack has some degree of orthogonal symmetry). Comparing these results with the radial ordering in the last column, a conclusion is that $R$ ordering is a more sensitive parameter in cylindrical packing experiments. The physical boundary (the wall effects) does influence the global structures deeply.

<table>
<thead>
<tr>
<th>Pack</th>
<th>Particle#</th>
<th>Global orders</th>
<th>Z ordering</th>
<th>R ordering</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$I_2$</td>
<td>$I_4$</td>
<td>$I_6$</td>
</tr>
<tr>
<td>C1</td>
<td>827</td>
<td>0.0200</td>
<td>0.0570</td>
<td>0.0475</td>
</tr>
<tr>
<td>C2</td>
<td>589</td>
<td>0.0156</td>
<td>0.0752</td>
<td>0.0549</td>
</tr>
<tr>
<td>C3</td>
<td>620</td>
<td>0.0263</td>
<td>0.0780</td>
<td>0.0535</td>
</tr>
<tr>
<td>C4</td>
<td>652</td>
<td>0.0321</td>
<td>0.1489</td>
<td>0.0365</td>
</tr>
<tr>
<td>C5</td>
<td>1258</td>
<td>0.0263</td>
<td>0.0568</td>
<td>0.0572</td>
</tr>
</tbody>
</table>

4.3.4 Particle Contact Number

To derive the contact numbers in experimental packings, a smaller number of internal particles were chosen to compute the average. The contact numbers versus pair-distance were computed because of the system uncertainties. The plots are shown in Figure 4.17. The plots show that as the packing density increases, the particle-particle contact number slowly increases.
Comparing these results with the results from the simulated and rough-walled experimental packings, a similar conclusion is made – the disordered experimental packs have a slightly smaller contact number (e.g. 5.7 in C5) than that of computer-simulated packings.
Combining all the collected structural information (density, RDFs, local and global ordering, contact number), a consistent conclusion is that cylindrical particles prefer a line- or face-contact to a point-contact in natural packs while computer simulations cannot capture this characteristic effectively, even though translational and rotational movements were constrained in the final stage during collective rearrangement. The computer simulations derived different results (density, ordering, and contact number) with experimental packs, but more or less close to the sphere packs, because of the point-contact logic, in which a (reordering) reorganization process is accompanied in the natural densification.

4.4 A Standard Packing Table

We end this section by building a standard packing table, which includes many different structural configurations. These packings will serve as a set of benchmarks for future standards to evaluate the ordering in arbitrary packings. Currently, the test packings include: nematic, cubatic, stacked logs, 2D/3D random, horizontal preferred, triplet cylinder bundles, fourth and sixth-folded, planar symmetry, lattice, concentric, etc. To simplify the analysis, we focus on packing structures disregarding any possible overlaps between cylindrical particles.

The different packing images are illustrated in Figure 18 as a sequence below: (a) 3D random lattice; (b) 2D random lattice; (c) cubatic lattice; (d) cubatic layered lattice; (e) fourth-folded planar; (f) sixth-folded lattice; (g) sixth-folded planar; (h) triplets; (i) tri-triplets; (j) cylindrical concentric; (k) cylindrical concentric with orthogonality; (l) 3d random slim cylinders; (m) nematic; (n) nearly horizontal; (o) horizontal; (p) horizontally stacked; (q) vertically stacked.
Figure 4.18  Imaginary different packing structures  
(fig. cont’d)
Table 4.12 lists the global ordering parameters for all the packings. From the table, we can see that all the parameters are small (close to zero) in 3D random packings (A and L); while in highly ordered parallel-stacked packings (P and Q), all the parameters have large values. These packings serve as the two extreme situations such that any other packings will have orientational parameters with values between them. For all of other test packing, there is at least one parameter that can characterize the packing structure, e.g. larger $M$ and $S_2$ and $I_2$ indicate a nematic or uniaxial structure; larger $S_4$ or $I_4$ values probably suggest a orthogonal or fourth-folded structure; smaller $S_{2,z}$ or larger $S_{4,z}$ indicates a near horizontal or planar structure, while larger $S_{2,z}$ indicates a vertical arrangement. More conclusions can be summarized below--
• M quantifies parallel (nematic) correlations. This parameter is weak (~zero) in 3D random structures and near unity for nearly nematic ones. Notice the parameter is close to 0.25 for planar structures.

• The combination of $S_{2,z}$ and $S_{4,z}$ can indicate packing structures with gravitational effects. Smaller $S_{2,z}$ and Larger $S_{4,z}$ suggest orthogonal structures, and larger $S_{2,z}$ tells vertical arrangement. Notice that $S_{2,z}$ is the smallest value of -0.5 and $S_{4,z}$ is a constant of 0.375 in horizontal arrangement.

Table 4.12 Global ordering parameters of different cylinder packings

<table>
<thead>
<tr>
<th>Packing</th>
<th>$M$</th>
<th>$S_{2,z}$</th>
<th>$S_{4,z}$</th>
<th>$S_2$</th>
<th>$S_4$</th>
<th>$I_2$</th>
<th>$I_4$</th>
<th>$I_6$</th>
<th># Particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2.99e-4</td>
<td>0.0243</td>
<td>-0.091</td>
<td>0.0354</td>
<td>-0.032</td>
<td>0.0409</td>
<td>0.111</td>
<td>0.0478</td>
<td>729</td>
</tr>
<tr>
<td>B</td>
<td>9.24e-4</td>
<td>0.0123</td>
<td>0.588</td>
<td>0.0339</td>
<td>0.274</td>
<td>0.0479</td>
<td>0.5915</td>
<td>0.1408</td>
<td>729</td>
</tr>
<tr>
<td>C</td>
<td>-4.9e-4</td>
<td>-0.029</td>
<td>0.571</td>
<td>0.0206</td>
<td>0.592</td>
<td>0.0297</td>
<td>0.7640</td>
<td>0.3487</td>
<td>729</td>
</tr>
<tr>
<td>D</td>
<td>-0.001</td>
<td>0</td>
<td>0.5833</td>
<td>0</td>
<td>0.583</td>
<td>5.7e-5</td>
<td>0.7638</td>
<td>0.3476</td>
<td>729</td>
</tr>
<tr>
<td>E</td>
<td>0.249</td>
<td>-0.5</td>
<td>0.375</td>
<td>0.2510</td>
<td>0.688</td>
<td>0.5</td>
<td>0.8292</td>
<td>0.5764</td>
<td>729</td>
</tr>
<tr>
<td>F</td>
<td>0.249</td>
<td>-0.5</td>
<td>0.375</td>
<td>0.2589</td>
<td>0.144</td>
<td>0.5</td>
<td>0.3752</td>
<td>0.7284</td>
<td>729</td>
</tr>
<tr>
<td>G</td>
<td>0.2490</td>
<td>-0.5</td>
<td>0.375</td>
<td>0.2500</td>
<td>0.1406</td>
<td>0.5</td>
<td>0.3750</td>
<td>0.7284</td>
<td>729</td>
</tr>
<tr>
<td>H</td>
<td>0.0076</td>
<td>-0.009</td>
<td>0.0923</td>
<td>0.0883</td>
<td>-0.005</td>
<td>0.1041</td>
<td>0.1409</td>
<td>0.0749</td>
<td>729</td>
</tr>
<tr>
<td>I</td>
<td>0.0098</td>
<td>-0.009</td>
<td>0.0923</td>
<td>0.0883</td>
<td>-0.005</td>
<td>0.1041</td>
<td>0.1409</td>
<td>0.0749</td>
<td>927</td>
</tr>
<tr>
<td>J</td>
<td>0.2488</td>
<td>-0.5</td>
<td>0.375</td>
<td>0.25</td>
<td>0.1406</td>
<td>0.5</td>
<td>0.375</td>
<td>0.3081</td>
<td>610</td>
</tr>
<tr>
<td>K</td>
<td>-0.001</td>
<td>-0.003</td>
<td>0.582</td>
<td>0.0167</td>
<td>0.2238</td>
<td>0.0177</td>
<td>0.5821</td>
<td>0.1246</td>
<td>610</td>
</tr>
<tr>
<td>L</td>
<td>-5.7e-4</td>
<td>0.007</td>
<td>-0.066</td>
<td>0.0387</td>
<td>0.0078</td>
<td>0.0457</td>
<td>0.0864</td>
<td>0.0494</td>
<td>376</td>
</tr>
<tr>
<td>M</td>
<td>0.5831</td>
<td>0.7642</td>
<td>0.3939</td>
<td>0.7644</td>
<td>0.3941</td>
<td>0.7644</td>
<td>0.3945</td>
<td>0.1188</td>
<td>376</td>
</tr>
<tr>
<td>N</td>
<td>0.2055</td>
<td>-0.454</td>
<td>0.2732</td>
<td>0.2435</td>
<td>0.0124</td>
<td>0.4556</td>
<td>0.3144</td>
<td>0.1846</td>
<td>376</td>
</tr>
<tr>
<td>O</td>
<td>0.2560</td>
<td>-0.5</td>
<td>0.375</td>
<td>0.3272</td>
<td>0.1366</td>
<td>0.5079</td>
<td>0.3913</td>
<td>0.3172</td>
<td>376</td>
</tr>
<tr>
<td>P</td>
<td>1</td>
<td>-0.5</td>
<td>0.375</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.9832</td>
<td>105</td>
</tr>
<tr>
<td>Q</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.9832</td>
<td>100</td>
</tr>
</tbody>
</table>

• $S_2$ and $S_4$ parameters are borrowed from liquid crystal theory, which can probe for the preferred orientation (or the director). Larger $S_2$ corresponds to nematic structures and larger $S_4$ corresponds to orthogonal (or multi-axial symmetric)
structures. Notice that $S_2$ is finite (0.2~0.3) for even-distributed horizontal packings.

- The computation of spherical harmonics ($I_2$, $I_4$, $I_6$) is highly recommended. These formulas are elegant in that they contain rich symmetric structural information. The quadratic invariant combination is able to express uniaxial, biaxial, cubatic and higher symmetric structures, and it is also frame independent. From the table, it can tell that $I$-parameters are superior to $S$-parameters because more structural information is detected, e.g. B, J, K, N, O packings.

- Concentric packings show strong radial-ordering, and the additional results are: $S_{2,r}=1.0$, $S_{4,r}=1.0$ (for J packing) and $S_{2,r}=-8.197e-4$, $S_{4,r}=0.5830$ (for K packing). Because J is in perfect cylindrical symmetry, the radial-ordering parameters equal to one. As a fraction of the particles switch to orthogonal directions, $S_{2,r}$ is reduced to zero and $S_{4,r}$ stays finite.

- Notice that there is no difference between triplet (H) and tri-triplet (I) packings, because they are same from a macroscopic viewpoint. To discern their differences, it must refer to local ordering parameters.

Table 4.10 lists the local ordering parameters of all the packings. Again there are small ordering values in random packings (A and L) and larger ordering values of highly regular packings (P and Q). Local ordering parameters are used to express local neighboring structures, e.g. $S_2$ is to express parallel correlation, and $S_4$ is to express orthogonal correlation and $S_6$ for higher-order correlation. These phenomena have been displayed in the table. Local ordering parameters are able to differentiate packings H and L, and the values increase as more and more particles form a neighboring cluster. $S_2$
alone cannot distinguish nematic (M), tri-triplets (I) and concentric (J) packings, but with
the combination of $S_2$, $S_4$, $S_6$, it is possible to differentiate them. For the latter packings,
all three parameters are finite, while only $S_2$ is large in nematic packings. Similarly, only
$S_4$ indicates orthogonal or fourth-folded structures. Surprisingly, horizontal (O) or nearly
horizontal (N) structures have weak correlations for the local ordering parameters, which
suggest the complexity in discerning angle distributions between 3D and 2D random
structures. For special concentric structures, the larger values of parallel and orthogonal
correlations are not surprising.

**Final note:** since there is possible overlap between neighboring cylinders, the
definition of neighboring pair (by a rough criterion of minimal distance less than $D_{\text{particle}}$)
is not very strict. However, according to the definition, all the possible neighbors are
included in the computation. Hence, these results are still representative.

<table>
<thead>
<tr>
<th>Packing</th>
<th>Local $S_2$</th>
<th>Local $S_4$</th>
<th>Local $S_6$</th>
<th># Particles</th>
<th>Filename</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2.393e-4</td>
<td>7.069e-3</td>
<td>-2.591e-4</td>
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<td>random latt</td>
</tr>
<tr>
<td>B</td>
<td>1.092e-2</td>
<td>0.3448</td>
<td>2.703e-2</td>
<td>729</td>
<td>random xy</td>
</tr>
<tr>
<td>C</td>
<td>1.843e-2</td>
<td><strong>0.5910</strong></td>
<td>0.1411</td>
<td>729</td>
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</tr>
<tr>
<td>D</td>
<td>7.517e-2</td>
<td><strong>0.6147</strong></td>
<td>0.1908</td>
<td>729</td>
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</tr>
<tr>
<td>E</td>
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<td>0.2777</td>
<td>729</td>
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</tr>
<tr>
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<td>0.1480</td>
<td><strong>0.5527</strong></td>
<td>729</td>
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</tr>
<tr>
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<td>0.2669</td>
<td>0.1599</td>
<td><strong>0.5589</strong></td>
<td>729</td>
<td>sixfold</td>
</tr>
<tr>
<td>H</td>
<td>0.1887</td>
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<td>309</td>
<td>triplets</td>
</tr>
<tr>
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<td><strong>0.4233</strong></td>
<td><strong>0.4965</strong></td>
<td><strong>0.4119</strong></td>
<td>927</td>
<td>tri-triplets</td>
</tr>
<tr>
<td>J</td>
<td><strong>0.8283</strong></td>
<td><strong>0.5849</strong></td>
<td>0.3750</td>
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<td>concentric2</td>
</tr>
<tr>
<td>K</td>
<td>2.669e-2</td>
<td><strong>0.4168</strong></td>
<td>6.582e-2</td>
<td>610</td>
<td>centric3</td>
</tr>
<tr>
<td>L</td>
<td>-9.479e-2</td>
<td>4.727e-3</td>
<td>8.402e-4</td>
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<td>random10</td>
</tr>
<tr>
<td>M</td>
<td><strong>0.5199</strong></td>
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<td>3.882e-2</td>
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</tr>
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</tr>
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</tr>
<tr>
<td>Q</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>100</td>
<td>stacked5</td>
</tr>
</tbody>
</table>

**Note:** gap=$D_{\text{particle}}$
CHAPTER 5
NETWORK STRUCTURE IN RANDOM CYLINDER PACKINGS

Network modeling is an important tool for handling complicate flow problems in porous media. The reasons are that the fundamental topological and morphological information of porous materials can be expressed in a network structure, and microscopic phenomena can be easily integrated into the networking modeling. Further, it can be operated at different scale because of the simplification of flow equations (using first principles). Therefore, it has been widely used by microscale flow simulators to explore different transport phenomena.

In network modeling, the pore space is expressed as a series of interconnected pores and pore-throats. The pores and their connections provide a complete geometrical and topological description of a porous material, thus giving the possibility of microscale fluid flow simulation. In this chapter, network structures were extract from random cylinder packings.

5.1 Network Structure

Numerous parameters associated with a network model can be divided into three groups: pore parameters (which include pore location, pore volume and the largest inscribed radius), pore-throat parameters (which include throat location, length, cross-sectional area, maximum inscribed radius, surface area, and hydraulic conductivity) and interconnectivity (throat index, neighboring pore indices and periodicity). Parameters and associated variable types in a general network are displayed in Table 5.1.
Table 5.1  All network parameters associated with a pore network

<table>
<thead>
<tr>
<th>Class</th>
<th>Variable name</th>
<th>Array name</th>
<th>Type</th>
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</thead>
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<td>Domain dimension</td>
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<td>Vector</td>
</tr>
<tr>
<td></td>
<td>Solid volume fraction</td>
<td>φ</td>
<td>Scalar</td>
</tr>
<tr>
<td>Pore</td>
<td>Pore #</td>
<td>NPORES</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Location</td>
<td>PLM</td>
<td>Vector</td>
</tr>
<tr>
<td></td>
<td>Volume</td>
<td>PVM</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Inscribed radius</td>
<td>RCPM</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Connectivity index</td>
<td>ICI</td>
<td>Scalar</td>
</tr>
<tr>
<td>Pore-throat</td>
<td>Throat #</td>
<td>NTHROATS</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Throat Length</td>
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<td>Vector</td>
</tr>
<tr>
<td></td>
<td>Neighbor pore index</td>
<td>ICM</td>
<td>Vector</td>
</tr>
<tr>
<td></td>
<td>Cross-sectional area</td>
<td>CSAM</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Surface area</td>
<td>SURFAM</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Inscribed radius</td>
<td>RCTM</td>
<td>Scalar</td>
</tr>
<tr>
<td></td>
<td>Hydraulic conductivity</td>
<td>HCM</td>
<td>Scalar</td>
</tr>
</tbody>
</table>

5.2 Numerical Technique

The computational domain is a unit cube with periodicity. This definition is consistent with the previous one, which was used to generate random dense packings. There are a total of 26 periodic computational domains that adjoin the primary domain in space. The advantages of using periodic domains are to eliminate wall effects and to build larger domains easily.

Pore locations in a network are defined as a set of points in the primary domain, each of which represents a stable position for an expanded spherical balloon. Mathematically, these positions are local maxima of the void-phase distance function, which is defined as the minimal distance from a point in void phase to any cylinder surface. Furthermore, these maximal inscribed spheres are constrained by at least four points on the surfaces of neighboring cylinders. Due to the sharp transitions of cylindrical surfaces, the total number of the local maxima can be large. Therefore, the numerical technique to find all pore locations is time-consuming. However, since most pores have
large overlaps, it is reasonable to merge smaller ones that are largely inside of large ones without compromising essential void phase properties. To find the local maxima, geometric simplex optimization is used here – a small tetrahedron undergoes continuous flips, extensions, and shrinkages to obtain the final optimal point. The starting points are regular lattice points in the domain, and the searching process ends when a certain stopping criterion is satisfied (e.g. the search coefficient is smaller than 1.0E-5, which is defined as the ratio of total number of optimal points located and total lattice points at one level). Once a pore location is known, the largest inscribed radius can be derived from the distance function. The pore concept is illustrated in Figure 5.1.

![Illustrations of pores in random cylinder packing](image)

**Figure 5.1** Illustrations of pores in random cylinder packing

In the network model, pores are uniquely defined to discretize the continuous void phase. That is, the summation of all pore volumes is equal to the total volume of void phase, and two pores are clearly separable. However, inscribed sphere centers in cylinder packings can be very close. The high frequency of this phenomenon suggests the open nature of random cylinder packings (at least at high $L/D$), which complicates the pore-discretization process. Therefore, certain merging criteria must be adopted before computing the pore volumes. In this work, the smaller pores were merged into larger
neighbors whenever their centers are inside a larger pore. This criterion is somewhat arbitrary, because most pore spheres remain overlapped with others (though less than those that were merged). Since overlapping pores are counted in the pore volume repeatedly and some pore space is not contained in the inscribed spheres, an adjustable scaling parameter is introduced to match the total void volume. The procedure we adopted is: (1) an initial pore volume is the summation of a group of conical frusta volumes along all its connecting throats; (2) the pore volume is rescaled to match the known total volume. This procedure not only makes the sum of the individual pore volumes identical to the total void-phase volume, but also emphasizes on the contribution from their throats, which is important in random cylinder packings with open, cage-like geometries.

Flow paths in a network form a map of the flow network. Flow paths are located deepest in the porous media (Lunchnicov et al., 1999), which usually have the lowest resistance to fluid flow (e.g. in Poiseuille flow, the flow rate is proportional to the fourth power of the radius in a pipe). An ideal flow path is defined in a ‘balloon-dragging’ process. Imagine an automatically expanding balloon (that keeps a spherical shape), which can move (without friction) in the pore space. The optimal positions, corresponding to local maxima in balloon diameter, are pore centers. If the balloon is dragged from a pore center to its neighboring pore center, it will be forced to shrink and then expand. The orbit of the balloon center is a flow path, which is generally an irregular curved line. Several mathematical attributes exist to define the flow path: there exist three equally minimal distances from three points on three nearby cylinder surfaces; these three
points form an acute triangle; the balloon passes through trumpet-shaped region with two
enlarged ends open to two pore centers (Figure 5.2).

![Figure 5.2 Illustration of pore-throat and flow path](image.png)

There is the difficulty in deriving these flow paths theoretically — a flow path is
formed by surface boundaries, so it is irregular. There are also several possible paths in
sparse porous media, while the one with minimal deviation from the straight line is
correct one. Here the Powell optimization was introduced to realize this process
numerically. The objective function is defined

$$\Psi = \min \sum_{i=1}^{9} (\psi_3(x_i, y_i, z_i) - \psi_1(x_i, y_i, z_i))^2$$

(5-1)

where $\psi_i$ is a set of minimal distance functions from a point to the cylinder surfaces.
Minimizing the summation of the square between the 1$^{\text{st}}$ and 3$^{\text{rd}}$ minimal difference
approximates tri-equal-distance flow path. There are total of nine points along each path,
which form a second-order polynomial in space. Using nine points along a flow path is a
compromise based on numerical limitations. The nine points $(x_i, y_i, z_i)$ are chosen as

$$x_i = a_0 + a_1 t_i + a_2 t_i^2$$

$$y_i = b_0 + b_1 t_i + b_2 t_i^2$$

$$z_i = c_0 + c_1 t_i + c_2 t_i^2$$

$t_i \in [0,1]$ and $i = 1...9$
In practice, the nine points are evenly distributed along $t_i$ from 0 to 1. Notice that when $t_i$ equals to 0 or 1, two fixed points that are the two connecting pore centers are obtained. Therefore, certain constraints on the constant coefficients are known in advance. Because a very small step is defined in locating the new conjugate direction during the Powell algorithm, allowing this method to converge to the minimal deviation.

Computation time is the major concern in the Powell optimization process. In small packings, a ‘brute-force’ technique is adopted to check all pores in the neighboring regions of a path. However, a Delaunay tessellation or similar technique should be used to pre-examine the neighboring pores and limit the number to a reasonable value before Powell optimization in order to save computation time, especially when the packing becomes large.

Once flow paths have been defined, other throat parameters are readily derived. The trumpet-shaped throat (flow channel) was represented by a series of conical frusta (Bryant et al., 1993) and was further approximated by a series of concentric cylinders (Zhang et al., 2005). The throat is divided into a set of connected cylinders of varying diameters, with three contacts lines along their surfaces. Three points at any cross section form an acute triangle. Therefore, a ‘triangular array’ model (Jackson & James, 1986, Higdon & Ford, 1996) is referred to compute throat parameters. The throat length ($L_{thr}$) is obtained from a summation of all ten conical frusta heights. The throat location is one of the nine points along a flow path with the minimal distance function ($r_{thr}$). The corresponding cross-sectional area (CSA) can be computed using the following formula (see Figure 5.3)
The surface area (SA) is the summation of the partial surface areas of contacting cylinders. The calculation is based on the triangular model, which emphasizes contribution of the local surfaces in the flow channel to the surface area. Note that rescaling is necessary to match the total cylinder surface area in the model.

\[
SA = \sum_{i=1}^{9} S_i = \sum_{i=1}^{9} L_i (\pi r_{fib})
\]

(5-4)

The remaining throat parameters are related to the concept of local solid volume fraction (SVF or \(\phi_{loc}\)), which can also be derived by the triangular array model. The local \(\phi\) is the ratio of cylinder-occupied areas that are inside of the computational triangular area and the whole triangular area (see Figure 5.3).

\[
\phi_{loc} = \frac{2\pi}{3\sqrt{3}} \left( \frac{r_{fib}}{r_{fib} + r_{loc}} \right)^2
\]

(5-5)

To compute the throat hydraulic conductivity, we use the procedure described by Thompson (2002) who developed local technique based on Jackson & James (1982,
1986) global approach. Specifically, different formulas are used for low vs. high solid volume fraction in the cylinder packings. For low $\phi$, the asymptotic expression for Stokes drag along parallel cylinders arranged is used on triangular array (Drummond & Tahir, 1984). The concept is applied in every throat locally to derive local permeability. The local permeability can be easily transformed into the local hydraulic conductivity via a simple relationship. This process is applied to every component (cylinder/frustum) of a throat, which gives ten local hydraulic conductivities that reflect the flow resistance along a flow path. Finally, we use the harmonic mean to calculate the throat hydraulic conductivity ($G_{thr}$). The whole procedure can be expressed by the following formulas:

\[
\frac{k_i}{r_{fib}} = \frac{1}{4\phi_i} (-\ln \phi_i - 1.498 + 2\phi_i - \frac{\phi_i^2}{2} + O(\phi_i^6)) \quad (5-6)
\]

\[
g_i = \frac{k_i A_i}{L_i} \quad (5-7)
\]

\[
\frac{1}{G_{thr}} = \sum_{i=1}^{10} \frac{1}{g_i} \quad (5-8)
\]

For high $\phi$, the parallel array model is not correct because more and more cylinder end-surfaces contribute to the boundary of flow channels. The flat end-surfaces and sharp surface transitions disturb the smooth streamlines, which influences the momentum transport process. In these cases, the pore-throats have a converging-diverging characteristic and a more duct-like geometry. Therefore, we use an effective hydraulic radius to compute the throat conductivity. Specifically, the effective hydraulic radius ($R_{eff}$) is the arithmetic mean of the equivalent radius ($r_{eq}$) and throat radius ($r_{thr}$), while the equivalent diameter ($D_{eq} = 2r_{eq}$) is four times the ratio of the throat volume (composed of
ten conical frusta volumes) and the throat surface area. Hydraulic conductivity is then calculated from the effective hydraulic radius (and throat length) using the Poiseuille equation.

\[ D_{eq} = 2r_{eq} = 4 \frac{V_{thr}}{S_{A_{thr}}} \quad (5-9) \]

\[ R_{eff} = \left( r_{eq} + r_{thr} \right) / 2 \quad \text{and} \quad R_{eff} = D_{eq} / 2 \quad (5-10) \]

\[ G_{thr} = \frac{\pi R_{eff}^4}{8 L_{thr}} \quad (5-11) \]

We end this section by mentioning interconnectivity in the network, which characterizes the connections among pores. To identify a true connection, we use three criteria: a flow path cannot pass through a cylinder (solid phase); a flow path cannot pass through a pore (other than the two it connects); a flow path has only one minimum in the distance function. These three criteria sufficiently exclude any false interconnections. However, due to the polynomial approximation of the flow path, we change the third criterion such that the distance function of the nine points on the path cannot be larger than that of the two ends (which are local maxima of pore centers).

- Algorithm

Our overall computational algorithm can be summarized as below:

1. Locate pore sites using the simplex method and obtain their maximal inscribed radii;

2. Merge pores according to the merging criteria;

3. Determine preliminary interconnectivity using a Delaunay tessellation (currently, we use a brute-force method in small packings);
4. Map flow paths using Powell optimization;
5. Calculate throat parameters (RCTM, CSAM, SURFAM, TLNM) and assign pore volumes (PVM);
6. Compute throat conductivities (HCM).

- Packings

Four simulated random close packings with different particle aspect ratios were used for network structural analysis (Figure 5.4).

Figure 5.4  Illustrations of random dense packings with varied cylinder aspect ratios
($L/D = 1, 5, 10$ and 20)

A detailed flowchart of the computational algorithm for network structure generations is displayed in Figure 5.5.
Figure 5.5  A flowchart of computer algorithm of network extraction
5.3 Computational Results

Four computer-simulated RCPs (with different cylinder aspect ratios, $L/D = 1, 5, 10$ and 20) are used to display the network results. Qualitative visualizations of these RCPs and the derived network interconnectivities are illustrated in Figure 5.4 and Figure 5.6. The network parameters are displayed in Table 5.2. In the following discussion, we highlight statistical properties of the derived networks.

Figure 5.6  Illustration of the network connectivity in four random close packings ($L/D = 1, 5, 10$ and 20)
5.3.1 Pore-size distribution (PSD)

Pore sizes here refer to the diameters of maximal inscribed spheres in a network. They are important as they are related to the local capillary pressure curves (Thompson, 2002). Pore volumes, which characterize the nature of void space in porous media, also influence the microscale flow and transport properties.

Figure 5.7 displays the histograms of the maximal inscribed radii in four packings. The sizes of maximal inscribed spheres are not uniform or symmetric distributions, but skewed. After mathematical manipulations, they can be fitted into a family of generalized Gamma distributions. The normalized pore size in Figure 5.8 is a dimensionless ratio of pore radii to the averaged cylinder radius in a packing. The normalized PSD displays several unique characteristics of the void structure in random cylinder packings: the distribution is narrow for equilateral cylinder packings, with an averaged inscribed pore size less than half of the cylinder size; As the cylinder aspect ratio increases, the distributions become wider and the averaged pore sizes become larger than the fiber size (e.g. for \(L/D > 10\), 20). Furthermore, the pore sizes can span several times the cylinder size, which reflects the heterogeneous character of the void phase in random cylinder packings.

The PSD characterizes the porous structure from a set of fixed points (inscribed spheres) in the void phase, while the pore volume distributions can characterize the void phase in another way. Figure 5.9 displays the histograms of the pore volume distributions in four packings. The pore volumes are not classical gamma distributions, but a special case of \(\gamma \leq 1\). The distribution shapes are far from \(\Gamma\)-like (or Weibull-like) but approach exponential, and their shape parameters (\(\gamma\)) decrease as the particles become slender. The
decrease in the average and variance of the pore volume tells that void phase has been divided more evenly by the slender particles. While the intrinsic reasons for these differences are not clear, part of the reasons maybe associated with the number of pores and the merging criterion (that is, pores in the network remain overlapping).

5.3.2 Throat size distributions (TSD)

Throat size refers to the diameter of a minimal inscribed sphere along a flow path, or the maximal sphere that can pass through. Throat size is a dominating parameter for determining friction along a flow channel in viscous flow (and thus influences the bulk permeability). Therefore, throat size information is important in many bond-controlled processes such as drainage.

A throat size was assigned as the minimal distance function (among nine points) along a flow path. Therefore, it is an approximation due to numerical limitations. However, it is meaningful because all possible neighbors surrounding a pore have been explored, and it is likely for a pore to connect another pore in a more direct manner. The interconnectivity structures are shown in Figure 5.6.

Figure 5.10 displays the histograms of throat size (or inscribed bottle-neck radii) distributions for four packings. The distributions are Γ-like (or Weibull-like), similar to the maximal inscribed pore radii distributions. This similarity is also found in other statistical properties, such as the mean of throat size. The average throat size steadily increases as the particles become slender, while the ratio of pore to throat is approximately two in all four packings. Surprisingly, this constant was found in other unconsolidated porous media (Payatakes et al., 1980; Jerauld & Salter, 1990; Hilpert et al., 2003). The normalized bottleneck radii were also fitted into a set of gamma
distributions (Figure 5.11), and the reverse behavior of shape parameters ($\gamma$) was found. That is, the shape parameters increase as cylinders become slender. Hence, the right-hand tail is less pronounced and closer to symmetric. The average throat size slowly increases, and reaches $\sim 0.6D_{\text{particle}}$ in the packing of $L/D=10$ and $\sim 1.0D_{\text{particle}}$ in the packing of $L/D = 20$.

Throat-length distributions in networks are examined to help characterize the heterogeneity in the void phase of random cylinder packings. A throat length here represents the orbit distance of the balloon center, which is approximated as ten line segments in the numerical scenario. This parameter incorporates curvature of a flow path and reflects the porous geometry.

Figure 5.12 displays the histograms of throat length distributions in four packings. The distributions are not similar in shape as PSD or TSD. The average throat length in the four packings decreases as cylinder aspect ratio increases, reflecting the fact that more pore centers are formed in the packing of slender cylinders. However, some throat lengths can span several pore distances, which is an unsatisfactory result of the current method. The reasons are the inability to discretize the whole void space by pores (that is, spherical pores only occupy part of the void phase), leaving a significant space for longer pores to be connected as long as their distance is smaller than the numerical limit (set as cylinder length).

The throat parameters (such as lengths, cross-sectional areas, surface areas) are used to compute each throat’s conductivity. This throat conductivity dramatically simplifies the fluid mechanics, and is a key factor to determine the correctness of resistor network computations. The throat conductivity is calculated from the local geometry and
integrated all throat parameters, so this approach is superior to purely stochastic methods for the assignment of conductivities.

5.3.3 Interconnectivity

The interconnectivity or the coordination number \( Z \) is a fundamental topologic metric for porous media. It is the mean number of bonds meeting at a node. This concept can be easily understood for some regular lattices (e.g. \( Z=6 \) for a simple cubic structure and \( Z=8 \) for a body-centered cubic structure). For unconsolidated random sphere packings, this number has been estimated to be 5.54 (Al-Raoush et al., 2005). For three-dimensional randomly placed points, the Voronoi tessellation gives a coordination number of approximate 15.5. This number provides an upper limit for our problem, since the domain is partly occupied by cylinders, which would reduce connectivity.

Qualitatively, we expect a smaller coordination number for thick cylinders (higher density packings), and a larger coordination number for slender cylinders (lower density packings).

The computational results for coordination numbers in different packings are displayed in Figure 5.13. From these histograms, it is clear that the coordination number increases as cylinders become slender. The increase in coordination number suggests a transition from tunnel-like to cage-like geometry locally. The computational results reflect this trend: the averaged coordination number is 5.3 in equilateral packing \( (L/D =1) \), and increases to 6.1 in the packing of \( L/D =10 \). The maximum interconnectivity also increases from 14 to 24.

We finish this part by mentioning the distribution of coordination numbers. The distribution in a network is very wide (e.g. the coordination number is 0–14 in equilateral
cylinder packing and 0–24 in \( L/D=10 \) cylinder packing). The wide distribution is indicative of the varied interconnectivity at a node. This characteristic suggests that the void structure in random cylinder packings is far different than artificial lattices with fixed coordination numbers (even they are heterogeneous). The existence of zero interconnectivity is of concern. The reason could be smaller pores involving unusual lower porosity locally, which correspond to the probable sites for gas trapping.

5.4 Conclusion

A novel approach for network modeling of dynamic flow problems in prototype fibrous materials is proposed, which were modeled as random packings of rigid cylinders. In the procedure of network extraction, the continuous irregular void phase is effectively transformed into pores, pore-throats, and a description of interconnectivity. In the network, pores are defined as a set of inscribed spheres, and throats are defined by the maximal spheres that can pass between two neighboring pores. The pore-scale network approach can integrate fundamental microscale phenomena (e.g. contact angle, film flow, snap-off etc.) and can be scaled up to study continuum behavior, so it is a highly promising method for future research.

Pore-size and throat-size distributions in the networks are skewed, and can be fit to a family of generalized gamma distributions. When normalized to the fiber size, the pore sizes vary more than the throat size. The ratio of the averaged pore size and throat size is about 2 in all four random dense packings. In addition, the pore volumes in the networks were calculated from their connecting pore-throat geometry, and their distributions were not \( \Gamma \)–like, but close to exponential.
The interconnectivity inside the networks is larger than 5, which is the approximate value for sphere packings. It was found that as the particle aspect ratio increases, the maximal packing density decreases and the interconnectivity increases. The larger interconnectivities reflect the open, cage-like nature of the fibrous materials, which is a characteristic of the real porous geometry.

Our research results show that some pore sites are the probable sites for gas trapping. Some numerical limitations exist in the network generation process. In the future, we will continue refining our algorithm to express the network more precisely. At the same time, we will adapt the network models for dynamic simulations to study transport parameters such as permeability, capillary pressure, and saturation curves.
Figure 5.7  Histograms of maximal inscribed radii distributions in four packings

Figure 5.8  Normalized maximal inscribed radii distributions in four packings
Figure 5.9  Histograms of pore volume distributions in four packings
\((L/D =1, 5, 10 \text{ and } 20)\)
Figure 5.10  Histograms of throat size distributions in four packings

Figure 5.11  Gamma distributions of normalized throat size in four packings
Figure 5.12  Histograms of throat length distributions in four packings
Figure 5.13  Histograms of coordination number distributions in four packings
Table 5.2 Some network parameters in four random close packings

<table>
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<tr>
<th>Packing</th>
<th>#cylinders</th>
<th>#pores (before merge)</th>
<th>#pores (after merge)</th>
<th>Normalized pore size</th>
<th>#throats</th>
<th>Normalized throat size</th>
<th>connectivity</th>
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<td></td>
<td>min</td>
<td>max</td>
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Table 5.3 Fitted PSD parameters in Gamma distributions

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<th>Location parameter (µ)</th>
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Table 5.4 Fitted TSD parameters in Gamma distributions

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CHAPTER 6
FLOW SIMULATIONS

Once the data for the network has been derived, several types of simulations (depending on the complexity of the real physical processes) can be performed, which include single-phase flow, two-phase flow, three-phase flow, non-Newtonian flow etc. In this chapter, we report single-phase flow simulations and compute the prototype material permeabilities. We also give a mathematical description of microscale physics inside a pore, and formulate the algorithm for dynamic multiphase flow preliminarily. Detailed simulations will be conducted in the future.

6.1 Single-phase Flow

6.1.1 Model Formulation

Single-phase flow implies only one fluid phase is present in the network. In a network model, pore-throats transport mass into and out of each pore and cause resistance. It is assumed that there is no accumulation of mass within the pore. Therefore, for constant density fluids, the mass balance in each pore (e.g. pore \( i \)) can be expressed conveniently in term of volumetric flow rate:

\[ \sum_j q_{ij} = 0 \]  

(6-1)

where \( j \) represents pore-throats connecting to pore \( i \). For low-Reynolds-number Newtonian flow, the volumetric flow rates (between pore \( i \) and pore \( j \)) can be written in terms of a single morphologic parameter (the hydraulic conductivity, \( G_{ij} \)) of the connecting pore-throat, even for complex throat geometries (Thompson, 2002).

\[ q_{ij} = \frac{G_{ij}}{\mu} (p_i - p_j) \]  

(6-2)
Two equations above can be written in a compact form (for pore \( i \)).

\[
\sum_j \frac{g_{ij}}{\mu} (p_i - p_j) = 0
\]  

(6-3)

Written above equation in each pore, a group of coupled equations are derived. For usual constant pressure conditions, some simple equations involving inlet and outlet pore pressures are added. For the boundary conditions of inlet constant flow rate and outlet constant pressure, additional equations involving flow rates are added into the system (Equation 6-4). The resulting equations are sufficient to solve for local pressures inside all pores. The local pressures can be used to determine the local flow rate (in each pore-throat).

\[
\sum_{j=\text{inlet}} q_{ij} = Q_{in}
\]  

(6-4)

For any other types of fluids such as Power Law, Bingham, Herschel-Bulkley etc., the corresponding various non-linear expressions for flow rate can be used (Balhoff & Thompson, 2003).

6.1.2 Numerical Procedure

The computational algorithm includes two steps: computing pore pressures and calculating the permeability components. In the first step, a set of linear equations is solved to derive all pore pressures. In the second step, the flow rate inside each pore-throat is calculated from Equation 6-2, and the flow rate of each pore is derived. The total volumetric flow rate is calculated by accumulating of all inlet pores. Finally the permeability is computed from flow rate, cross-sectional area and length, viscosity and pressure difference.
During the simulation, we use periodic boundary conditions to avoid errors caused by a small characteristic domain size. A simple flow chart of the algorithm is given in Figure 6.1.

**Figure 6.1** A flowchart of single-phase flow simulation
6.1.3 Permeability Results

- Isotropic Packings

A series of prototype fibrous materials with varied aspect ratio fibers \((L/D=1, 5, 10, 20)\) were studied, with a density range of 65%-20%. Good agreement was observed when compared these computational results with the extensive experimental permeability data of different real fibers (from Jackson & James, 1986). Table 6.1 summarizes the computational permeability results for different isotropic packings. For easy comparison, the results were written in the dimensionless form of \(K/a^2\), where \(a\) is fiber radius and \(K_{\text{avg}}\) is the average value from three orientational tensor components. In addition, the maximal percent differences among three components were computed and listed in the last column.

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<th>(K_{\text{avg}}/a^2)</th>
<th>(K_{xx}/a^2)</th>
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<tr>
<td>10</td>
<td>0.25</td>
<td>0.1339</td>
<td>0.1470</td>
<td>0.1228</td>
<td>0.1319</td>
<td>9.8%</td>
</tr>
</tbody>
</table>

Note: \(\text{max diff} = \max (K_i-K_{\text{avg}})/K_{\text{avg}}\)

In general, the results validate the network approach as an effective technique to model fibrous material permeability. Specifically, the following conclusions can be made: Firstly, the derived permeability tensors are close to isotropic, which confirms the randomness of the cylinder packings; Secondly, the averaged permeability steadily increases as the fiber aspect ratio increases, which is caused by the sparser nature of the random packings. Thirdly, since the local porosity concept was introduced in calculating
throat conductivities (based on derived asymptotic formulas (Drummond & Tahir, 1984; Jackson & James, 1986)), it is quite reasonable to extend this approach to higher porosity fibrous materials. On the other hand, for low porosity packings, the duct-like Poiseuille approach can be used. Therefore, this modeling approach is quite general.

The numerical results from single-phase-flow simulations, together with extensive experimental results for fibrous materials from Jackson & James (1986), are displayed in Figure 6.2. The numerical results match the measured permeabilities for real fibrous materials, giving us a confidence of the network approach.

![Predicted permeabilities of prototype fiber packings](image)

Figure 6.2  Predicted permeabilities of prototype fiber packings

- **Anisotropic Packings**

  For anisotropic materials, anisotropic physical properties (e.g. permeability) are expected. Can the network model effectively capture this property?

  Table 6.2 displays the permeability results for different anisotropic packings from our simulations. For highly regular packings of nearly nematic cylinders, in which fibers...
have a preferred z orientation, permeability anisotropy does appear. There are \( \sim 24\% \) \((\phi=0.35)\) and \( \sim 59\% \) \((\phi=0.45)\) difference in their orientational components. While in another anisotropic packing of highly horizontal preference (but uniformly distributed in x-y plane), the anisotropy is not very sensitive. Only \( \sim 7\% \) \((\phi=0.35,\) equal density of 3D random packing\) difference among their orientational components in the permeability tensor is observed. When the packing density increases to \( \phi=0.45\), the difference increases to 25\%. Comparing these results with the isotropic packing \((L/D=10,\) SVF=0.35\) in table 6.1, we find that the scalar value of component-averaged permeability increases as the structural randomness decreases. That is, the permeability is lowest in the isotropic case, higher in the preferred horizontal case, and the highest in the nearly nematic case. Although all the scalar components are increasing, the component in the priority orientation increases faster.

<table>
<thead>
<tr>
<th>Packing</th>
<th>SVF</th>
<th>(K_{avg}/a^2)</th>
<th>(K_{xx}/a^2)</th>
<th>(K_{yy}/a^2)</th>
<th>(K_{zz}/a^2)</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nema10_35</td>
<td>0.35</td>
<td>0.09498</td>
<td>0.08603</td>
<td>0.0880</td>
<td>0.1109</td>
<td>24.11%</td>
</tr>
<tr>
<td>Nema10_45</td>
<td>0.45</td>
<td>0.2302e-1</td>
<td>0.1781e-1</td>
<td>0.1989e-1</td>
<td>0.3139e-1</td>
<td>58.99%</td>
</tr>
<tr>
<td>Tenth10_35</td>
<td>0.35</td>
<td>0.7604e-1</td>
<td>0.7923e-1</td>
<td>0.7515e-1</td>
<td>0.7373e-1</td>
<td>7.23%</td>
</tr>
<tr>
<td>Tenth10_40</td>
<td>0.40</td>
<td>0.3665e-1</td>
<td>0.3923e-1</td>
<td>0.3998e-1</td>
<td>0.3075 e-1</td>
<td>25.18%</td>
</tr>
</tbody>
</table>

Note: Difference=\((K_{max}-K_{min})/K_{avg}\)

6.2 Two-phase Flow

6.2.1 Dynamic Imbibition Formulation

In dynamic displacement, capillary forces exist because of the interface between the two phases. General assumptions during simulation are the following: (1) creeping flow; (2)
no gravity (due to small scale). Based on the microscopic flow phenomena and pore morphology, a tentative model formation is formulated below.

The constitutive equation for each phase is given by

\[ q_{ij,w} = \frac{G_{ij,w}}{\mu_w}(P_{w,j} - P_{w,i}) \]  (6-5)

\[ q_{ij,nw} = \frac{G_{ij,nw}}{\mu_{nw}}(P_{nw,i} - P_{nw,j}) \]  (6-6)

The parameters are subscripted with \( w \) or \( nw \) to denote the wetting and nonwetting phases respectively. In practice, the conductivities are further defined as \( G_{ij,w} = g_{w,ij}G_{ij} \) and \( G_{ij,nw} = g_{nw,ij}G_{ij} \). This approach is analogous to using relative permeability as a nondimensional coefficient on absolute permeability to express multiphase permeability at the continuum scale. For dynamic imbibition problem, unsteady mass balance on pore \( i \) gives

\[ \frac{V_i}{\Delta t}(S_{w,j}^{l+1} - S_{w,i}^l) = \sum_j g_{w,ij}G_{ij} \left( P_{w,j} - P_{w,i} \right) \]  (6-7)

\[ \frac{V_i}{\Delta t}(S_{nw,i}^{l+1} - S_{nw,i}^l) = \sum_j g_{nw,ij}G_{ij} \left( P_{nw,j} - P_{nw,i} \right) \]  (6-8)

where \( V_i \) is the volume of pore \( i \) and \( S_i \) is the phase saturation (nondimensional) in pore \( i \).

The superscripts \( l \) and \( l+1 \) denote the current and future times in the simulations as dictated by the time step \( \Delta t \). The time indexes are left off on the right-hand-side terms to allow the equations to remain general for either explicit or implicit solution, which will be discussed in next section.

Equations (6-6) and (6-7) are tied together through two auxiliary equations: the first is the definition of phase saturation, and the second is the capillary pressure inside pore \( i \).
Note that the capillary pressure \( P_c \) is the point-wise capillary pressure that applies at a local interface as opposed to the spatially averaged capillary pressure used in continuum-scale simulations. Hence, it is subscripted with the pore index to indicate that its value will differ from one pore to another.

Finally, the boundary conditions can be constant pressure or flow rate of inlet pores and the usual constant pressure of outlet pores.

\[
p_i = \text{constant (or } \sum_j q_j = \text{constant)} \quad \text{for inlet pores} \tag{6-11}
\]

\[
p_i = \text{constant} \quad \text{for outlet pores} \tag{6-12}
\]

### 6.2.2 Proposed Numerical Procedure

- **Explicit Method**

  The fully explicit method would involve evaluating all right-hand-side terms in Equations at the current time level, which means each equation can be solved explicitly for \( S^{t+1} \). Subsequently, new pressures would be obtained through the appropriate constitutive equations (i.e. capillary pressure versus saturation relationships). In numerical implementation, this method might be unstable.

- **Implicit Method**

  For the fully implicit method, all terms that have \( P \) or \( S \) functionality are evaluated at the forward timestep. Making this change in notation and substituting Equations (6-6) and (6-7) to eliminate \( P_{nw} \) and \( S_{nw} \) leads to the following pair of coupled equations.
These equations can be solved using the multidimensional Newton-Raphson method, which involves iterating on the Matrix equation \([J]{\delta x} = \{-f\}\). The Jacobian contains the partial derivatives \(\partial f/\partial S_w\) and \(\partial f/\partial P_w\), and \(\{\delta x\}\) represents updates for the current values of \(S_w\) and \(P_w\). Because of the expense of creating the Jacobian, quasi-Newton methods can be used that approximate the Jacobian at each timestep using information from previous iterations.

### 6.2.3 Additional Considerations

The relative conductances \(g_{ij,w}\) and \(g_{ij,nw}\) are easy to understand, but they are difficult to quantify. In practice, a pore-throat cannot have a small relative conductance to the nonwetting phase. The throat is either closed to the nonwetting phase (if it has not yet been invaded) or it has a high value of relative conductance (near 1.0) once the nonwetting phase has invaded. Hence, the relative conductance function might show a discontinuous behavior.

The pore capillary pressure function is also complicated. It has a minimum at a finite value of capillary pressure, with an unstable branch to the right (higher value of \(S_w\)). A qualitative sketch is shown in Figure 2.8. The capillary pressure curve is also hysteretic and depends on the local interface configuration (e.g. the order in which adjacent throats have invaded during drainage or imbibition). For the model to be
physically realistic, it is essential that local pore saturations should not have values along
the unstable branch except during rapid transients. Currently, it is unclear whether
numerical solution techniques will naturally obey this stability constraint.
CHAPTER 7
CONCLUSIONS AND FUTURE RESEARCH DIRECTIONS

7.1 Conclusions

We have studied random cylinder packings using both experiments and computer simulations. In the experimental part, equilateral cylinders were used, and a series of densification experiments were performed in a cylindrical container and in a rough-walled cubic container. The detailed packing structures (e.g. packing density, positional ordering, orientational ordering and wall effects) were analyzed using a state-of-the-art X-ray microtomography technique. In the numerical simulations, a variety of different cylinder packings were created using a collective rearrangement algorithm. The 3d random close packing limits were explored for different particle aspect ratios. The resulting packing structures from two methods were compared. The void space in cylinder packings was studied numerically, and the geometrical and topological information was expressed as a pore network. The statistical properties of the derived networks were analyzed. Finally, single-phase-flow simulation was performed on the pore networks, and the corresponding permeabilities were computed. The derived permeability data match extensive experimental results from different fibrous materials, thus giving confidence in the network modeling approach.

The major conclusions of this work includes: (a) we explored 3D random dense packing limits numerically of cylindrical particles with sharp surface transitions (while other investigators studied experimentally, or theoretically, or using particle substitute such as spherocylinders to avoid the numerical challenge); (b) we analyzed packing structures, real packing density and contact numbers in experimental packs using X-ray
microtomography technique and developed a numerical way to reconstruct particle-scale packings; (c) we quantified the randomness in varied disordered cylinder packings and built several benchmarks in quantifying the packing structures, which extend far from the crystalline state; (d) we realized the network approach in sparse fibrous materials, and predicted material permeability by single-phase-flow modeling; (d) we proposed the network modeling frame for multiphase flow simulation in the future.

The numerical contributions include: (a) cylinder-cylinder contact and random packing algorithms; (b) quantification of the orientational ordering in random packings; (c) connected-component algorithm in reconstructing particle-scale packings; (d) network generation algorithm; (e) 3D visualization technique in Fortran-Matlab interface.

7.2 Future Research Directions

7.2.1 Packing

In generating 3D random cylinder packings, computation time is a major limit in numerical process. This problem is especially apparent when the packing becomes dense and approaches its maximum density limit, where particle movements are largely blocked. This problem also occurs when the initial structure (e.g. fiber positions) are not uniform in space. Therefore, our packing models are limited to a number range of several hundred to thousand cylinders. In engineering applications, it may be necessary to generate very large packings such as hundreds of thousands or millions of particles. The current algorithm cannot be used in this context. To accelerate the modeling process, a simpler method to compute contact criteria is needed, which works in the random structures relevant to these simulations.
In modeling real fibrous materials, curved cylinders and 2D random fiber mats (widely used) are two major considerations to improve the applicability of the modeling.

7.2.2 Structure

In quantifying the positional order, the particle RDF is based on the distance from a particle center to the reference particle surface. This RDF choice weakens the influence of positional ordering (for slender particles) compared to the RDF derived from the minimal distance between two particle surfaces. The peer work involving spherocylinders from Williams and Philipse (2003) suggests that second choice is better. The computation will relate to the averaged excluded volume of two cylinders statistically. See Onsager (1949) and Blaak et al. (1999) for future references.

7.2.3 Network

In the process of expressing the void space into a pore-throat network, it seems that the pore merging process influences the statistical results. Qualitatively, if we allow more pores to be merged, the pore shape and volume, throat length and the connectivity are altered. Therefore, the merging criterion is somewhat arbitrary. The irregularity of pore shapes influence microscopic film flow and the snap-off phenomenon. Therefore, comparison and selection of pore-merging criteria is an important future topic.

From the geometry and topology of a network, many statistical properties can be produced. However, single variable (or marginal) distributions (e.g. PSD, TSD and CND) cannot characterize the complexity of real porous media statistically. For completeness and a better basis for statistical models, we should study pairwise and spatial correlations of these variables. Future students can refer to Bryant et al. (1993) for the spatial
orrelations and Lindquist (2001) for joint distributions, starting from the present network structures.

7.2.4 Simulation

For simulation, multiphase flow is certainly an important topic. By simulating the dynamic displacement process, several transport parameters (e.g. capillary pressure versus saturation, relative permeability versus saturation, residual saturation) could be predicted. In manufacturing functional composites, these simulations could also be used to predict void information in the bulk materials.

Furthermore, upscaling and streamline-scale simulations are also future challenging topics.
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