

A STRATIFIED MODEL OF A RANDOM BED OF EQUAL-DIAMETER SPHERES CONFINED BY A PLANE

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A mathematical model has been formulated of a bed of equal-diameter spheres randomly dumped on a plane surface. Based on this model relationships have been derived for the area porosity of the bed as a function of the distance from the supporting plane. The model contains three parameters with well defined physical meaning and its asymptote for infinite distance from the supporting plane satisfies criteria typical for random beds of spheres.

Random systems of spheres, or more generally particles, have been subject of interest of several scientific disciplines such as *e.g.* statistical thermodynamics, research of the structure of gels, suspensions or aerosols, chemical engineering, chemical technology, *etc.*

Random beds of spheres serve as models of non-crystalline structure and molecular aggregates. A widely used method of the study the structure of random packings of spheres have been computer simulations making use of *e.g.* the Bernal model with various functions describing the interatomic potential. The computer then searches for those configurations minimizing the potential energy of the system¹.

Another computer-aided approach consists of the simulation of the process of slow settling of a thin system of spheres. This type of computer simulation, which is essentially a statistical-geometrical approach (the spheres must not overlap and must be supported), has been used, for instance, by Tory and coworkers² to study the probability density distribution function of porosity in a spherical volume. The results of these authors of the distribution of nearest neighbours yield as most probable six points of contact. They also found a significant anisotropy, *i.e.* differences in results valid for a horizontal and vertical plane, appearing due to the effect of gravity forces.

A review of the extensive literature on the structure of a packing of spheres have been presented by Haughey and Beveridge³.

The properties of random beds of spheres, particles of irregular shape and mixture of such particles have been studied theoretically and experimentally by Debbas and Rumpf⁴. Based on statistical properties of the packing these authors regard a given bed of equal-diameter spheres as random provided it satisfies the following criteria: 1) The area porosity in all cross sections is identical; 2) The area porosity equals the volume porosity of the whole bed; 3) The mean area of discs appearing on a given plane cut through the layer equals $\pi d^2/6$; 4) The frequency function of the diameters of discs, Φ , appearing on a given plane cut through the bed is given by $z(\Phi) = \Phi/(d(a^2 - \Phi^2)^{1/2})$; 5) The cumulative distribution function of diameters

of these discs is $Z(\Phi) = 1 - (d^2 - \Phi^2)^{1/2}$. The experimental study of these authors has shown the minimum volume porosity satisfying still the criteria of randomness to be about 0.35. Below this limit the beds were losing their random character judging from the viewpoint of the above listed criteria. A tendency to regularities, or the deviations from the random properties exhibit packings subjected to vibrations. The most compact packing exhibited porosity in certain region as low as 0.316; the minimum porosity 0.26, however, could not be reached owing to the effects of the walls and flaws of the shape of the spheres.

A statistical-geometrical argument has been used by Gotoh and Finney⁵ to construct a most probable tetrahedron and to calculate the overall packing density, taking the latter to be a system of tetrahedron aggregates of spheres.

Scott⁶ observed experimentally that porosity of packings in solid containers ranges between two well-defined limits. In his measurements the results were corrected by extrapolation to a vessel of infinite diameter and length. Mild vibrations resulted in the so-called "dense random packings" while filling the vessel by sliding the packing over an inclined plane lead to "loose random packings". The extrapolated porosities for these two random packings amounted to 0.36 (6) and 0.40 (9). This phenomenon was further studied by Bernal and Mason⁷.

A distribution function for a randomly distributed set of spheres as well as for a mixture of spheres was investigated by Herczynski⁸. The minimum porosity of a two-component mixture depends generally on the diameter of both types of spheres and their relative concentration. Numerical experiments have shown that for the sphere diameter ratio between 1 and 2 and the relative concentration between 0.1 and 0.9 the minimum porosity changes little.

Zagrafskaya and coworkers⁹ have applied a globular model for a theoretical and experimental study of colloid systems. They start from random partly organized systems composed of individual spheres and chains of spheres. The probability density distribution function of the number of contacts has the normal distribution with the mean 5.9 for individual spheres; after vibrations this mean increases to 7.1. The experimental data of these authors furnish the following correlation between the mean number of contacts, \bar{n} , and the porosity of the bed in the form $\bar{n} = 2.62/\epsilon$. These authors worked out also a correlation for the mean pore radius within the bed, r_p , in the form: $r_p/r = 0.62\epsilon/(1 - \epsilon)$, valid in the interval $4 \leq \bar{n} \leq 10$.

Packings of spheres, or particles of other shapes, formed by dumping into a container exhibit deviations from random configuration in the proximity of container walls. This phenomenon has been studied experimentally in a number of papers¹⁰⁻¹³. Most of these papers deal with the effect of cylindrical walls and the course of porosity near such walls. The significance of such studies rests in the effects on the flows of both gases and liquids such as these exist in packed-bed type separation units, catalytic reactors, gas-solid reactors, heat recuperators, etc. The peculiarities in the course of porosity in the proximity of the walls results in anomalous gas flows¹⁴ in this region, by-passing and in case of trickle beds channelling and wall flow formation¹⁵.

Analogously there exists the effect of horizontal plane surfaces confining the packing such as e.g. supporting grids, etc. This phenomenon, however, has been so far little studied. Indirectly, though its existence has been recognized in the experimental techniques of the study of radial porosity profiles when the horizontal sections adhering directly to the bottom and the top are eliminated. Experimentally the course of porosity in the vertical direction in the proximity of a plane surface has been studied by Benenati and Brosilow¹¹. Their results revealed a course similar to radial profiles with characteristic oscillations reaching 4.5 sphere diameter deep into the packing.

In this paper a mathematical model has been formulated enabling description of the packing of equal-diameter spheres confined by a plane. The parameters of this

model relate to well-defined physical quantities characterizing the bed and at infinite distance from the plane the model satisfies the criteria for a random bed set by Debbas and Rumpf⁴.

THEORETICAL

Already a mere visual observation of packing of equal-diameter spheres resting on a flat support through the wall of a glass container reveals that the influence of the pad reaches several sphere diameters deep into the bed. In a bed of equal-diameter sphere the first "layer" has all spheres with their centers one sphere radius away from the supporting plane. The obviousness of the arrangement of the spheres into "layers" grows weaker with increasing distance from the plane until it completely disappears in the random structure of the bed.

It is assumed that the packing may be divided along its whole height into horizontal layers parallel to the supporting plane while each layer contains the same number of spheres, N . The situation is schematically shown in Fig. 1, where the shadowed area depicts the supporting pad. The total thickness of the first layer is $(r + \delta)$ and may be divided by a plane designated by the index $i = 1$ at a distance r from the plane into two parts. The plane $i = 1$ is the geometrical locus of all centers of all N spheres belonging into the first layer; some of these spheres are shown in Fig. 1 and designated by capital letters A, B and C. Depending on the compactness of the bed these spheres may either contact (*e.g.* spheres A and B) or be mutually separated as *e.g.* spheres B and C and C.

Immediately above the first layer there are additional layers, each 2δ thick, divided by horizontal plane referred to by the indices $i = 2, 3, 4, \dots$, into two, this time equal, parts. These planes are thus planes of symmetry of corresponding layers and, in addition, geometrical loci of most probable position of centers of N spheres belonging to each layer. The probability density distribution function of the centers of spheres is symmetrical with respect to the planes i and is shown below in Eq. (7). Some of the

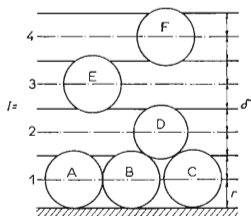


FIG. 1

Sketch of a Random Bed of Spheres Resting on a Plane Surface

Shaded area represents supporting plane surface.

spheres with the most probable position of center in layers $i = 2, 3, 4$ are shown in Fig. 1 by letters D, E and F. The limits between two neighbouring layers are horizontal planes at the position depending on the index i : $(r + (2i - 3)\delta)$ (Excepting $i = 1$) and $(r + (2i - 1)\delta)$. Maximum spacing of centers of two spheres belonging to the same layer is thus 2δ .

The distribution function of the probability density for the position of centers of the spheres in the i -th layer $f_i(x^i)$, where x^i is the coordinate of the distance of center of the sphere from the plane i , shall be expressed in the form

$$f_i(x^i) = 1/(2\delta) + (1/\delta) \sum_n \cos(\pi n x^i/\delta) \exp[-(\pi n)^2 D h_i/\delta^2]. \quad (1)$$

In this formula h_i designates the distance of the plane of maximum probable position of centers of spheres in layer i from the plane $i = 1$.

The function $f_i(x^i) = f_i$ satisfies the differential equation

$$\partial f_i/\partial h_i = D \partial^2 f_i/\partial (x^i)^2 \quad (2)$$

typical for random, diffusional processes whose solution on an infinite interval gives the normal distribution function.

The distribution function (1) is valid on the interval $-\delta \leq x^i \leq \delta$ and satisfies the condition of symmetry with respect to the plane i

$$\partial f_i/\partial x^i = 0 \quad \text{for} \quad x^i = 0. \quad (3)$$

Further we have that

$$\int_{-\delta}^{\delta} f_i dx^i = 1. \quad (4)$$

On expressing the quantity h_i by means of the characteristic scale of the layer

$$h_i = (i - 1) 2\delta \quad (5)$$

and the quantity D , characterizing the scatter of the spheres about the plane i , in terms of the characteristic radius of the spheres

$$D = \eta 2r \quad (6)$$

we obtain

$$f_i(x^i) = 1/(2\delta) + (1/\delta) \sum_n \cos(\pi n x^i/\delta) \exp[-(\pi n)^2 4\eta(i - 1)/(\delta/r)]. \quad (7)$$

From additional properties of this function there follows that for $i = 1$ the Dirac function results with the non-zero value at the point $x^1 = 0$, which, indeed, is the required property of the first layer.

The dimensionless variance of position of centers of spheres in a general layer is given by:

$$\begin{aligned} \sigma^2 &= \int_{-\delta}^{+\delta} f_i(x^1) (x^1/\delta)^2 dx^1 = \\ &= (1/2) + \sum_n 4(-1)^n \exp [-(\pi n)^2 4\eta(i-1)/(\delta/r)] / (\pi n)^2 \end{aligned} \quad (8)$$

and in the special case of $i = 1$ we obtain $\sigma^2 = 0$ while for $i \rightarrow \infty$ then $\sigma^2 = 1/3$.

In the limiting case of infinite distance from the supporting plane the distribution of centers of the spheres is given by

$$\lim_{i \rightarrow \infty} f_i = 1/(2\delta). \quad (9)$$

This result is identical with the assumption (4) of Debbas and Rumpf⁴ concerning the properties of a random packing of spheres.

If the packing of spheres is confined in the radial direction by a cylindrical surface of radius R , then the quantity $(1 - \epsilon)$ in a general plane parallel to the supporting plane equals the mean square radius of discs appearing after hypothetical sectioning of the bed by this plane, multiplied by the dimensionless group $N(r/R)^2$.

Consider now a plane alpha parallel to the plane (i) at a distance x_0^i in the positive direction (*i.e.* away from the support). Provided the position of alpha satisfies the inequality

$$r - \delta \leq x_0^i \leq \delta$$

then alpha intersects only spheres belonging to the layers (i) and ($i + 1$). The dimensionless mean square radius of discs on sectioned spheres belonging to the layer (i) is then given by the integral

$$I^i(x_0^i) = \int_{x_1^i = x_0^i - r}^{\delta} f_i(x^1) [1 - ((x_0^i - x^1)/r)^2] dx^1. \quad (10)$$

The mean square radius of discs on spheres of layer ($i + 1$) sectioned by the plane alpha is

$$I^{i+1}(x_0^i) = \int_{-\delta}^{x_0^i + r - 2} f^{i+1}(x^{i+1}) [1 - ((x_0^i - 2\delta - x^{i+1})/r)^2] dx^{i+1}. \quad (11)$$

After transformation

$$x_0^{i+1} = x_0^i - 2\delta. \quad (12)$$

Eq. (11) leads to

$$I^{i+1}(x_0^{i+1}) = \int_{-\delta}^{x_0^{i+1}+r} f^{i+1}(x^{i+1}) [1 - ((x_0^{i+1} - x^{i+1})/r)^2] dx^{i+1} \quad (13)$$

where x_0^{i+1} designates position of alpha with respect to the plane $(i+1)$.

In case that the position of the plane alpha satisfies the inequality

$$0 \leq x_0^i \leq r - \delta$$

alpha sections spheres belonging to the layers $(i-1)$, (i) , $(i+1)$. The dimensionless square radius of the discs on sectioned spheres belonging to (i) is

$$I^i(x_0^i) = \int_{-\delta}^{\delta} f^i(x^i) [1 - ((x_0^i - x^i)/r)^2] dx^i. \quad (14)$$

Similarly for the discs belonging to the layer $(i+1)$

$$I^{i+1}(x_0^i) = \int_{-\delta}^{x_0^{i+1}+r-2\delta} f^{i+1}(x^{i+1}) [1 - ((x_0^i - 2\delta - x^{i+1})/r)^2] dx^{i+1}. \quad (15)$$

Finally the mean square radius of discs belonging to the layer $(i-1)$ is given by

$$I^{i-1}(x_0^i) = \int_{2\delta+x_0^{i-1}-r}^{\delta} f^{i-1}(x^{i-1}) [1 - ((x_0^i + 2\delta - x^{i-1})/r)^2] dx^{i-1}. \quad (16)$$

By transformation (12) the integral (15) yields

$$I^{i+1}(x_0^{i+1}) = \int_{-\delta}^{x_0^{i+1}+r} f^{i+1}(x^{i+1}) [1 - ((x_0^{i+1} - x^{i+1})/r)^2] dx^{i+1} \quad (17)$$

and by transformation

$$x_0^{i-1} = x_0^i + 2\delta \quad (18)$$

integral (16) changes to

$$I^{i-1}(x_0^{i-1}) = \int_{x_0^{i-1}-r}^{\delta} f^{i-1}(x^{i-1}) [1 - ((x_0^{i-1} - x^{i-1})/r)^2] dx^{i-1}. \quad (19)$$

After these transformations all integrals take generally the form

$$I^m(x_0^m) = \int f^m(x^m) [1 - ((x_0^m - x^m)/r)^2] dx^m, \quad (20)$$

where

$$m = (i - 1), i, (i + 1).$$

For those cases, when the plane α appears below the plane (i) , i.e. if x_0^i are negative, corresponding integral can be written directly. For the region

$$-\delta \leq x_0^i \leq \delta - r$$

the contribution of the layer (i) is integrated in the limits $\langle -\delta; r + x_0^i \rangle$ and the contribution of the layer $(i - 1)$ in the limits $\langle x_0^{i-1} - r; \delta \rangle$.

In region

$$\delta - r \leq x_0^i \leq 0$$

spheres of three layers contribute. The contribution of the layer (i) is obtained after integration following Eq. (20) in the limits $\langle -\delta; \delta \rangle$, the contribution of the layer $(i - 1)$ after integration in the limits $\langle x_0^{i-1} - r; \delta \rangle$ and the contribution of the layer $(i + 1)$ in the limits $\langle -\delta; x_0^{i+1} + r \rangle$.

On substituting appropriate distribution functions, integration and on reverse transformation using Eqs (12) and (18) all integrals relate to the common frame of reference referred to the plane (i) . On designating the dimensionless mean square radius of discs on all sectioned spheres by I , the area porosity may be expressed by

$$\varepsilon = 1 - N(r/R)^2 I. \quad (21)$$

Introducing for brevity a new dimensionless coordinate

$$x = x_0^i/\delta \quad (22)$$

the dimensionless mean square radius of discs on all spheres sectioned by plane at a position x satisfying

$$1 - (r/\delta) \leq x \leq (r/\delta) - 1 \quad (23)$$

may be expressed by

$$\begin{aligned} I &= I^i(x) + I^{i+1}(x) + I^{i-1}(x) = \\ &= 2r/(3\delta) - 4(\delta/r)^2 \sum_n \exp(-P(n, i)) (-1)^n / (\pi n)^2 - \end{aligned}$$

$$\begin{aligned}
& - 2(\delta/r) \sum_n \exp(-P(n, i+1)) \cos(\pi n(x + (r/\delta)))/(\pi n)^2 - \\
& - 2(\delta/r)^2 (x-1) \sum_n \exp(-P(n, i+1)) (-1)^n/(\pi n)^2 + \\
& + 2(\delta/r)^2 \sum_n \exp(-P(n, i+1)) \sin(\pi n(x + (r/\delta)))/(\pi n)^3 + \\
& + 2(\delta/r)^2 (x+1) \sum_n \exp(-P(n, i-1)) (-1)^n/(\pi n)^2 - \\
& - 2(\delta/r) \sum_n \exp(-P(n, i-1)) \cos(\pi n(x - (r/\delta)))/(\pi n)^2 - \\
& - 2(\delta/r)^2 \sum_n \exp(-P(n, i-1)) \sin(\pi n(x - (r/\delta)))/(\pi n)^3, \quad (24)
\end{aligned}$$

where

$$P(n, i+m) = (\pi n)^2 \eta(i-1+m)(r/\delta) \quad \text{for } m = -1, 0, 1. \quad (25)$$

In regions

$$(r/\delta) - 1 \leq x \leq 1 \quad (26)$$

$$-1 \leq x \leq 1 - (r/\delta) \quad (27)$$

the expression for I takes the following form

$$\begin{aligned}
I &= I^i(x) + I^{i\pm 1}(x) = \\
&= 2r/(3\delta) \pm 2(\delta/r)^2 (x \pm 1) \sum_n \exp(-P(n, i)) (-1)^n/(\pi n)^2 - \\
& - 2(\delta/r) \sum_n \exp(-P(n, i)) \cos(\pi n(x \pm (r/\delta)))/(\pi n)^2 \mp \\
& \mp 2(\delta/r)^2 \sum_n \exp(-P(n, i)) \sin(\pi n(x \mp (r/\delta)))/(\pi n)^3 \mp \\
& \mp 2(\delta/r)^2 (x \mp 1) \sum_n \exp(-P(n, i \pm 1)) (-1)^n/(\pi n)^2 - \\
& - 2(\delta/r) \sum_n \exp(-P(n, i \pm 1)) \cos(\pi n(x \pm (r/\delta)))/(\pi n)^2 \pm \\
& \pm 2(\delta/r)^2 \sum_n \exp(-P(n, i \pm 1)) \sin(\pi n(x \pm (r/\delta)))/(\pi n)^3, \quad (28)
\end{aligned}$$

where in the case of double signs the upper holds for the case of inequality (26), the lower for the case when the inequality (27) applies.

The expressions (24), (28) may be regarded as generally valid for $i > 2$. For the layer $i = 2$, however, it holds only in region $x \geq (r/\delta) - 1$. In their remaining part of the layer $i = 2$ and in the whole layer $i = 1$ there are terms in the expressions (24) and (28) relating to the nonexisting layer $i = 0$. This drawback, however, can be

circumvented formally by introducing the layer $i = 0$ with a zero probability density throughout the layer

$$f_0(x^0) = 0 \quad \text{for all } x^0. \quad (29)$$

For the purpose of Eqs (24) and (28) we shall extend the definition (25) by

$$P(n, 0) = \infty. \quad (30)$$

For practical calculations of the porosity profiles in regions influenced by the spheres of the first layer ($i = 1$) it is though preferable to derive special relations in which we use for $i = 1$ directly the Dirac Function

$$\begin{aligned} f_1(x^1) &= \infty & \text{for } x^1 &= 0 \\ &= 0 & \text{for } x^1 &\neq 0 \end{aligned} \quad (31)$$

instead of its harmonic expansion (7). This improves the rate of convergence of the appropriate series at the costs, of course, of more complicated computational algorithm.

The expression valid in region immediately adhering to the supporting plane is

$$I = 1 - (\delta/r)^2 x^2, \quad \text{for } -(r/\delta) \leq x \leq 1 - (r/\delta). \quad (32)$$

DISCUSSION

The derived expression written in Eqs (21), (25) and (28) may be used to compute the profiles of area porosity in a random packing of equal-diameter spheres confined by a plane surface. The model contains a total of three parameters. The dimensionless damping coefficient η ; dimensionless group $N(r/R)^2$ and the dimensionless half-thickness of the model layer (δ/r) . The effect of these parameters on the shape of the profile is demonstrated in Fig. 2.

The parameter η characterizes the rate of damping of the visibly layered structure and the transition to the random structure. Packings with corresponding high value of the damping coefficient exhibit small "penetration depth" of the porosity oscillations. Graphically the differences at various dampings are demonstrated under otherwise identical conditions by curves 1 and 2 in Fig. 2. For higher of the selected values of the damping coefficient (curve 1) the depth of penetration of the oscillations amount to about 5 times the sphere diameter.

In the limit $R \rightarrow \infty$, and in practice for sufficiently large values of the ratio R/r when the packing is free of the effect of the container walls, the dimensionless group $N(r/R)^2$ takes a nonzero value independent of R . As follows from a geometrical argu-

ment the group is maximum for packings with maximum compactness. For a triangular configuration of spheres $N(r/R)^2 = \pi/(2\sqrt{3}) \approx 0.907$. On the contrary, the loosest possible configuration is with the spheres in the corners of a triangle with the fourth in the center missing. Then: $N(r/R)^2 = \pi/9 \approx 0.349$. As it may be expected that the scatter of centers of the spheres in the second layer is small, the dimensionless group $N(r/R)^2$ permits the porosity of the first minimum to be determined with sufficient accuracy from

$$\varepsilon_{\min} \doteq 1 - N(r/R)^2. \quad (33)$$

On the contrary, the expressions in Eqs (24) and (28) converge in the limit $i \rightarrow \infty$ to the same value. In a sufficient distance from the confining plane, in region free of the oscillations the porosity, ε_∞ , thus takes values given by

$$\varepsilon_\infty = 1 - N(r/R)^2 (2/3) (r/\delta). \quad (34)$$

Eq. (21) may be rearranged then to give

$$\varepsilon = 1 - (1 - \varepsilon_\infty) (3/2) (\delta/r) I, \quad (35)$$

where ε_∞ may be taken as a new parameter replacing $N(r/R)^2$. Graphically the effect of the dimensionless group $N(r/R)^2$ is illustrated in Fig. 2 by curves 3 and 4 under otherwise identical conditions. A greater number of spheres in the layer, N , in case of the curve 4 thus becomes manifest through the overall shift toward lower values of porosity while the curves 3 and 4 intersect only at the origin.

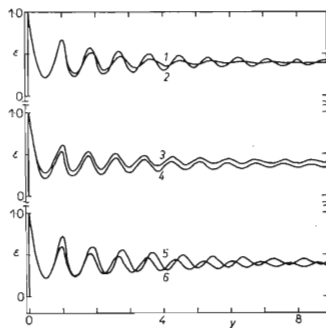


FIG. 2

Porosity Profiles Predicted by the Stratified Model

Curve: 1 $\eta = 0.01$; $N(r/R)^2 = 0.787$; $(\delta/r) = 0.88$; 2 0.005; 0.787; 0.88; 3 0.005; 0.721; 0.83; 4 0.003; 0.787; 0.85; 5 0.005; 0.787; 0.90; 6 0.005; 0.787; 0.85.

Physical meaning of the parameter (δ/r) is obvious: it is the half wave-length of the oscillations. Theoretically the lowest value of this parameter is obtained for the triangular arrangement of the spheres. Then $(\delta/r) = \sqrt{2}/2 \approx 0.707$. This configuration, however, is not random. The effect of (δ/r) is illustrated graphically in Fig. 2 by curves 5 and 6. Lower value of (δ/r) caused not only a different wave-length of the oscillations but also lower porosities in phase-corresponding positions.

The relations formulated on the basis of the proposed model are applicable to packings of approximately spherical compact particles of equal nominal size provided the equivalent diameter of the sphere is used.

The model may be applied also to porosity profiles in the proximity of cylindrical walls provided the ratio (r/R) is small.

LIST OF SYMBOLS

d	diameter of spheres forming the packing
$f_i(x^i), f_i$	probability density distribution function for the position of centers of spheres
h_i	distance of plane (i) from plane $i = 1$
l	mean square radius of all discs on sectioned spheres as a multiple of r^2
$l^i(\cdot), l^{i+1}(\cdot), l^{i-1}(\cdot)$	mean square radius of discs on sectioned spheres belonging to layer shown by the superscript expressed in terms of r^2
\bar{n}	mean number of contact points
n	summation index
N	number of spheres in a single layer of the stratified model
$P(n, \cdot)$	quantity defined by Eqs (27) and (31)
r	radius of spheres forming the packing
R	radius of cylinder containing the packing
x^i	coordinate of distance of centers of spheres within the layer with respect to plane (i)
$x_0^i, x_0^{i+1}, x_0^{i-1}$	coordinates of position of a given plane with respect to plane shown by superscripts
x	dimensionless coordinate of position of an arbitrary plane with respect to plane (i)
y	distance from confining plane measured in multiples of d
δ	half-thickness of model layer
Φ	radius of discs appearing on sectioned spheres
η	damping coefficient in the stratified model
ε	area porosity (function of position)
$\varepsilon_{\min}, \varepsilon_{\infty}$	porosity of the first minimum and porosity in region free of the oscillations

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