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# Functional regression-based fluid permeability prediction in monodisperse sphere packings from isotropic two-point correlation functions

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ABSTRACT

We study fluid permeability in random sphere packings consisting of impermeable monodisperse hard spheres. Several different pseudo-potential models are used to obtain varying degrees of microstructural heterogeneity. Systematically varying solid volume fraction and degree of heterogeneity, virtual screening of more than 10,000 material structures is performed, simulating fluid flow using a lattice Boltzmann framework and computing the permeability. We develop a well-performing functional regression model for permeability prediction based on using isotropic two-point correlation functions as microstructural descriptors. The performance is good over a large range of solid volume fractions and degrees of heterogeneity, and to our knowledge this is the first attempt at using two-point correlation functions as functional predictors in a nonparametric statistics/machine learning context for permeability prediction.


## 1. Introduction

In adsorption, filtration, separation, chromatography, and catalysis applications, understanding the impact of microstructural morphology of random, heterogeneous, porous materials on effective transport properties is key [1–5]. Indeed, establishing quantitative structure-property relationships and determining the importance of different three-dimensional morphological characteristics is a prerequisite for targeted optimization of a microstructure, and hence fine-tuning of a material for a specific purpose [6]. In e.g. gel chromatography, spherical gel particles are packed into a column through which solutes in liquid suspension flows. The separation of the solutes is determined largely by the flow around the gel particles, which is a function of microstructure.

There is generally a lack of good analytical expressions for quantitative structure-transport relationships [7], although there are empirical expressions like the Kozeny-Carman equation for estimating permeability [8,9] which can sometimes be rather useful. Exact prediction of transport properties, effective diffusivity and permeability, in random, heterogeneous, porous materials requires in principle complete knowledge of the microstructure in three dimensions i.e. the geometry of the solid-liquid interface. Complete microstructural information in three dimensions is not accessible for real materials; hence, there is a great interest in finding useful proxies i.e. pieces of limited microstructural information that capture the essential features.

A well-investigated set of microstructural descriptors are the $n$-point correlation (probability) functions for $n = 1, 2, \ldots$, introduced by Brown [10] for estimating effective transport properties in random materials. The set of all these correlation functions provide complete microstructural information, but are in practice increasingly unavailable for increasing $n$ [11]. Two-point and three-point correlation functions and microstructural parameters derived from them have been used to establish lower and upper bounds for effective transport coefficients. This includes both effective diffusivity (including other physical processes mathematically analogous to effective diffusivity, like electrical conductivity, thermal conductivity, and magnetic permeability) [12–18] and fluid permeability [19–24]. In two fairly recent papers [25,26], a microstructural parameter extracted from the three-point correlation function and introduced by Torquato [16] is used in prediction of effective diffusivity in monodisperse and polydisperse hard sphere systems.

In this work, we are concerned with permeability in random sphere packings consisting of monodisperse, impermeable, solid,

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hard spherical particles. This is a highly interesting case e.g. for the design of materials for chromatography, separation and catalysis. Several different pseudo-potential models are used to obtain varying degrees of microstructural heterogeneity i.e. varying the distribution of pore space sizes. Systematically varying solid volume fraction and degree of heterogeneity, virtual screening of more than 10,000 material structures is performed, simulating fluid flow using a lattice Boltzmann framework and computing the permeability. It is evident that the permeability will depend not only on the solid volume fraction (and on the specific surface which is perfectly correlated with the solid volume fraction in this setting) but also on the microstructure. We develop a functional regression model for permeability prediction based on using isotropic two-point correlation functions as microstructural descriptors. The performance is good over a large range of solid volume fractions and degrees of heterogeneity, and to our knowledge this is the first attempt at using two-point correlation functions as functional predictors in a nonparametric statistics/machine learning context for permeability prediction.

## 2. Results and discussion

### 2.1. Microstructure generation

Statistically homogenous and isotropic sphere packing microstructures are generated using a hard sphere monte carlo algorithm. A target solid volume fraction $\phi$ is chosen uniformly distributed in the range $0.10 \leqslant \phi \leqslant \phi_{\mathrm{RCP}}$, where $\phi_{\mathrm{RCP}} \approx 0.64$ is the random close packing limit [27]. Monodisperse spheres ($N = 4096$) with radii $R = 1$ are placed uniformly distributed in a cubic simulation domain $[0, L]^3$ with periodic boundary conditions, possibly with some overlaps. The initial simulation domain length $L$ is chosen such that the initial solid volume fraction (given that no particles overlap) is $\phi_{\mathrm{start}} = \min(\phi, 0.55)$, where $\phi$ is the target solid volume fraction. The reason for this choice is that it is increasingly more difficult to remove overlaps for increasing solid volume fractions, which will be further discussed below. The relation between solid volume fraction and simulation domain length is

$$L = \left(\frac{4\pi N}{3\phi}\right)^{1/3} R. \tag{1}$$

After the initialization, the system is relaxed i.e. all overlaps are removed by sequentially translating the spheres by normal distributed displacements with zero mean and standard deviation $\sigma_{\mathrm{t}}$. The amount of overlap is characterized by a system energy function,

$$E = \sum_{i=1}^{N}\sum_{j=i+1}^{N} \max\left(0, (R_i + R_j)^2 - \|\mathbf{x}_i - \mathbf{x}_j\|^2\right), \tag{2}$$

where $R_i = R_j = 1$ are the radii of spheres $i$ and $j$ and $\mathbf{x}_i$ and $\mathbf{x}_j$ are their positions. The energy between two non-overlapping particles is zero and the energy between two overlapping particles is a quadratic function of the amount of overlap. Only translations which lead to a decrease in $E$ are accepted. The standard deviation $\sigma_{\mathrm{t}}$ is initially 0.05 but is then adaptively adjusted in each sweep, striving for an acceptance probability target value of 0.25. After relaxing the system (i.e. after the system energy has reached $E = 0$, which is possible because the solid volume fraction is set to a value below the random close packing limit, hence all overlaps can be resolved), it is equilibrated by performing 100 sweeps at $E = 0$. If $\phi \leqslant 0.55$, we are now done with generating a basic structure and proceed to obtain controlled heterogeneities as described below. However, if $\phi > 0.55$, the simulation domain is gradually compressed in constant increments of $\phi, \Delta\phi = 5 \cdot 10^{-5}$, until the target solid volume fraction is reached, enforcing that $E = 0$ at each step before compressing further. The system is then equilibrated again by performing 100 sweeps at $E = 0$. All these steps are performed using the same algorithm for random translations as described above.

To obtain controlled heterogeneities, realizations of periodic random pseudo-potentials $f$ are generated and the spheres perform random displacements (with the constraint that $E = 0$) using the same algorithm as above until the total potential of the system,

$$P = \sum_{i=1}^{N} f(\mathbf{x}_i), \tag{3}$$

is minimized (it is possible that the optimization of $P$ converges to a local but not global minimum, i.e. a metastable state, but this fact does not compromise our purpose of generating these structures because the wide range of heterogeneities is nevertheless obtained). Three different types of pseudo-potentials are used: For type (I), we use a uniformly random number $M$ (between 1 and 128) of uniformly distributed attractive quadratic point potentials centered in the points $\mathbf{y}_m$,

$$f(\mathbf{x}) = -\sum_{m=1}^{M} \|\mathbf{x} - \mathbf{y}_m\|^{-2}. \tag{4}$$

For types (II) and (III), realizations of Gaussian random fields are simulated on a $128^3$ grid using fast Fourier transform methods [28]. For type (II), we use a covariance function family taken from Euclidean quantum field theory [28,29], with the density of the measure given by the Fourier transform of the covariance function being

$$\gamma(\mathbf{p}) = \left(1 + \left(p_1^{2k} + p_2^{2k} + p_3^{2k}\right)^l\right)^{-n} \tag{5}$$

for $k = 1, n = 1.765$, and $l = 1.5$. Briefly, the square root of this density is multiplied by independent white noise on a grid and inverse Fourier transformed to yield a random periodic function, the spatial scale of which is varied to generated different degrees of heterogeneity. For type (III), we use a Matérn covariance function family [30] with

$$\gamma(\mathbf{p}) = \frac{8\pi^{\frac{3}{2}}\Gamma\left(v + \frac{3}{2}\right)2^v v^v}{\Gamma(v) l^{2v}} \left(\frac{2v}{l^2} + 4\pi^2\left(p_1^2 + p_2^2 + p_3^2\right)\right)^{-v-\frac{3}{2}} \tag{6}$$

using $v = 5$ and $l = 0.2$, and $\Gamma$ is the gamma function. As for type (II), the spatial scale is varied to generate different degrees of heterogeneity. A considerable range of degrees of heterogeneity is hereby systematically explored. We refrain from getting into more technical detail about the Gaussian random fields, but proceed to showing examples of pseudo-potentials and the corresponding generated sphere packing microstructures in Fig. 1, showing one structure from each class, all with solid volume fraction $\phi = 0.15$. In total, in excess of 10,000 sphere packing microstructures are generated in this fashion. Data sets with both uniformly distributed, random solid volume fractions $\phi$ as well as fixed solid volume fractions $\phi = 0.10, 0.15, \ldots, 0.55$ are generated, the latter to study the impact of microstructure isolated from solid volume fraction and specific surface. The algorithms are implemented in Julia [31,32]. The computational time required to generate the structures is 0.6 h on average.

### 2.2. Flow simulations

The generated microstructures are converted to binary voxel structures of size $192^3$ voxels, that in turn are converted to geometric surface data (triangulated surfaces which is necessary for input into the flow simulation software; however, the flow simulations are performed on a voxel grid). The fluid flow through the