Multiscale modeling of fixed-bed reactors with porous (open-cell foam) non-spherical particles

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Multiscale modeling of fixed-bed reactors with porous (open-cell foam) non-spherical particles: Hydrodynamics

Saurish Das, N.G. Deen*, J.A.M. Kuipers

* Corresponding author.
E-mail address: N.G.Deen@TUE.nl (N.G. Deen).

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1. Introduction

Materials of interest such as catalysts are usually noble or transition metals. To synthesize the final catalyst, these catalytically active materials are usually deposited on so called catalyst carriers. The final catalyst can be in the shape of pellets, extrudates or structured, and in fixed-bed reactors these catalyst particles are usually packed randomly. Most catalytic processes depending on the type/shape of the catalysts...
employed and/or the reaction phases, are subjected to various degrees of mass transfer limitations. The reactions (often heterogeneous) take place at the surface of the catalyst carriers and are usually highly exothermic; and proper cooling is required to avoid thermal runaway. To overcome problems related to hot spot formation, multi-tubular packed bed reactors are preferably used, where a large number of parallel slender columns are placed inside a cooling jacket. The diameter of an individual reactor tube is small and is in the same order as that of the catalyst particle diameter. As a result, it provides efficient cooling of each individual tube, thereby suppressing severe radial temperature gradients.

The main aim of the present work to investigate the hydrodynamics in slender packed beds with particles/pellets consisting of open structures. These porous pellets/particles can be used to fill the reactor bed randomly. Optimum selectivity and performance demands higher flow rate inside the pellet, with minimum pressure drop. In this study, the porous particles are composed of open-cell solid foams, and different combinations of particle shape and column diameter are considered. Open-cell solid foams are novel materials which have been extensively used over the past few decades to form porous media for its excellent thermal and mechanical properties. Due to its large surface area to volume ratio, it has ample applications in heat transfer devices like heat exchangers, thermal energy absorbers, vaporizers, etc. Open-cell solid foams are also gaining popularity as catalyst support in several industrial catalytic processes, e.g. partial oxidation, alkylation, oxy-chlorination, hydrogenation, dehydrogenation etc. [1,2]. Instead of using a single block of porous foam structure, catalytic foam particles/pellet provide additional modularity for loading and unloading during maintenance, as-well-as during catalyst preparation [1,3]. Different combinations of particle size and column diameter can be used to form slender packed beds of different porosities using same foam sample supplied from the foam manufacturer.

In literature, there exists a large variety of numerical studies on packed bed reactors consisting of solid spherical particles [4-9] and a few for non-spherical particles [10-14]. For those studies, flow through the interstitial space between the particles is resolved and the characteristic length scale for the flow remains more or less the same throughout the bed. However, for the present problem, there exists two different hydrodynamic length scales, one at the micro-scale: internal pores in the porous particles and another one at the macro-scale: external voids generated by randomly packed particles of different shapes. As a result, it constitutes a problem of a bi-disperse porous media, where the fluid has two flow paths, through the foam particles (often referred to as rate of perfusion [15,16]) and around the particles. Perfusion offers the greatest benefits in fluid-solid interfacial exchange if fluid is forced through the porous structure creating a more homogeneous micro-environment, rather than just improving convection at the outer surface of the particles where the fluid has two flow paths, through the particles and around the particles. This distribution of the flow depends on both the distribution of the particles in the column and the internal resistance of the porous particles. The present flow situation is schematically depicted in Fig. 1. Numerically resolving both the scales is not computationally feasible and hence a multiscale modeling approach needs to be adopted. We develop an accurate finite volume (FV) method to simulate simultaneous flow through and around the non-spherical porous particles in a Cartesian computational domain. Volume averaging theory (VAT) [17] is adopted for macroscopic modeling of flow through the porous particles, whereas the classical Navier-Stokes equations are solved to fully resolve the flow through the interstitial space between the particles. In our model, a single set of equations is solved over the full domain for both the flow inside and outside the porous particles. The interface between the fluid and the porous medium is modelled as diffuse, where the continuity of velocity and viscous shear stress are satisfied across the interface. In VAT the flow variables are spatially averaged over a representative elementary volume (REV) of the homogeneous porous medium and the variation which occurs on a scale smaller than the REV needs to be modeled separately in the form of closures. Although some details of the flow field are lost due to averaging, the hierarchical modeling technique is essential when two substantially different length scales are persist in the problem. The drag closure (flow resistance) for the fluid-foam interaction at the micro-scale has been obtained separately by performing fully resolved direct numerical simulations using idealized geometry of a single unit-cell of open-cell solid foams [18]. The cylindrical wall effect is important for slender packed beds and in the adopted Cartesian computational domain it is resolved by an implicit, sharp interface immersed boundary method (IBM) [19]. A challenging task is to computationally generate random packings of non-spherical particles (rectangular or cubic shaped particles). The natural filling of the column with non-spherical particles is simulated with the aid of a glued-sphere discrete element method (DEM) [20].

This paper is organized as follows. First, we describe the numerical techniques and implementation of the current macroscopic modeling approach for our hydrodynamic study. Subsequently, the developed model is applied to a classical test case, flow through a channel partially filled with a porous medium; where the present results are compared with analytical results. The glued-sphere DEM model to generate random packings of non-spherical particles is described briefly as well. Packings are generated using three different sized particles in four columns of different diameters (particle to column diameter ratio \( \sim 3-40 \)) where the flow Reynolds number \( (Re_D) \), based on the particle size \( (d_p) \), is varied from creeping flow to \( Re_D \geq 400 \). After presenting the resulting packing structures in terms of porosity, particle distribution, particle orientation etc., flow simulation results are presented. Finally, we present the main conclusions of our work.

2. Multiscale modeling of fluid flow

To predict flow through a homogeneous porous medium, the majority of existing studies pertinent to moderate and high Reynolds number flows, use a Forchheimer-Darcy flow model (Forchheimer’s extension to Darcy’s law for high inertial flows). This model features a non-linear, 1-D momentum equation, which correlates the pressure gradient with the volumetric average velocity as:

\[
\frac{\mu}{K} \langle u \rangle + \frac{\mu_b}{K} \langle u \rangle \cdot \langle u \rangle = -\nabla \langle p \rangle
\]

where \( K \) is the permeability and \( b \) is the Forchheimer inertial coefficient. Both are a function of porosity \( (\epsilon) \) and geometry of the porous structure. Fluid viscosity and density are represented by \( \mu \) and \( \rho \), respectively, where \( \langle u \rangle \) represents the superficial velocity. For a bounded porous medium or porous media flow in a channel, the above model suffers from a well-know disadvantage in the sense that it cannot capture the presence of any wall [21]. Similarly, in case a fluid flows over and through a porous medium, it is not possible to match the continuity of velocity and shear stress at the fluid/porous medium interface using this model. To overcome this, Brinkman [22] proposed an effective viscous shear stress term as:

\[
\frac{\mu}{K} \langle u \rangle + \frac{\mu_b}{K} \langle u \rangle \cdot \langle u \rangle - \mu_{eff} \langle u \rangle^2 = -\nabla \langle p \rangle
\]

where \( \mu_{eff} \) is the effective viscosity which arises due to the presence of the porous medium. The value of \( \mu_{eff} \) is determined experimentally or by fully resolved simulation. In the existing literature either \( \mu_{eff} = \mu \) or \( \mu_{eff} = \mu/\epsilon \) has been used for the pseudo fluid inside the porous zone. On the contrary, its value has been shown to lie within a range of \( 5.1 < \mu_{eff}/\mu < 10.9 \) by Givler and Altabelli [23]. However, a thorough numerical study by Alazmi et al. [24] suggests that the value of \( \mu_{eff} \) has very negligible effects on overall macroscopic results.

For packed bed with solid particles, it is possible to apply directly the no-slip condition at the fluid-solid interface. For very simple flow
systems, where the fluid/porous-medium interface conforms to the computational grid, it is still possible to solve the flow inside and outside the porous block separately, in a decoupled manner by imposing continuity of shear stress and velocity at the interface [25]. However, the present problem involves a random packing of non-spherical porous particles and as such it creates a complex interstitial space between the particles where the surfaces of the particles are not always aligned along the grid lines. To overcome this problem, we adopt a volume averaging theory (VAT) based macroscopic modeling approach for the full bed, where a single set of equations is solved over the full domain and continuity of velocity and shear stress across the interface are enforce in an implicit way. Fig. 1 schematically shows the present multiscale modeling strategy, where: \( l_{\text{micro}} \), \( l_{\text{REV}} \), and \( l_{\text{macro}} \) represent the length scales for the micro-pores, the REV, and the macroscopic scale, respectively; where \( l_{\text{micro}} \ll l_{\text{macro}} \).

2.1. Governing equations and solution methodology

To derive volume-averaged Navier-Stokes equations, a certain quantity \((\phi)\) is decomposed into its interstitial average \((\langle\phi\rangle)\) and the spacial deviation/fluxuation \((\hat{\phi})\) as: \(\phi = \langle\phi\rangle + \hat{\phi}\). Generally, two different types of averaging are used in porous media literature, namely the super macroscopic average \((\langle\phi\rangle)\) and interstitial average \((\langle\phi\rangle)\), defined as,

\[
\langle\phi\rangle = \frac{1}{V_{\text{REV}}} \int_{V_{\text{REV}}} \phi \, dV \quad \text{and} \quad \langle\phi\rangle = \frac{1}{V_{\text{REV}}} \int_{V_{\text{REV}}} \phi \, dV
\]

(3)

where \(V_{\text{REV}}\) is the total volume of the REV, and \(V_{\text{REV},f}\) is the volume occupied by the fluid within the REV. These two averages are related by porosity \((\varepsilon \equiv V_{\text{REV},f}/V_{\text{REV}})\) as: \(\langle\phi\rangle = \varepsilon \langle\phi\rangle\). For a Newtonian, incompressible fluid, by integrating (spatial) the microscopic transport equations over the REV, the following volume-averaged (macroscopic) governing equations are obtained [17]:

\[
\nabla \cdot (\varepsilon (\mathbf{u})/\varepsilon) = 0
\]

(4)

\[
\rho \frac{\partial (\varepsilon (\mathbf{u})/\varepsilon)}{\partial t} + \rho V \cdot (\varepsilon (\mathbf{u})/\varepsilon) + \rho V \cdot (\varepsilon \mathbf{u}\mathbf{u}) = -\varepsilon \nabla \cdot (\mathbf{P}) + \nabla \cdot (\varepsilon \mathbf{P}) + \varepsilon \mathbf{g} + S_{\varepsilon} (\mathbf{u})
\]

(5)

where \(\mathbf{P}\) represents the viscous stress tensor \((\mathbf{P} = \mu \left[\nabla (\mathbf{u})/\varepsilon + (\nabla (\mathbf{u})/\varepsilon)^T\right]\)) and \(\nabla \cdot (\varepsilon \mathbf{u}\mathbf{u})\) the hydrodynamic dispersion which is insignificant as compared to the macroscopic inertial force and hence can be neglected [26,27]. To calculate the viscous stress tensor \((\mathbf{P})\) inside the porous medium, the interstitial velocities \((\mathbf{u})\) and \(\mu = \mu_0 \varepsilon\) are used. The momentum sink term, \(\beta (\mathbf{u})\), represents the flow resistance inside the porous medium. In this single field formulation for flow, the constant \(S\) acts as an indicator function to represent pure fluid cell \((S = 0)\) or a computational cell with porous structure \((0 < S \leq 1)\) (see Fig. 2). When it is a pure fluid cell, \(\varepsilon = 1\) and at the same time, \(S = 0\), as a result Eq. (5) reduces to the classical Navier-Stokes equations.

Before solving the above conservation equations, porosity values are assigned to each computational cell based on the particle location. The computational cells inside the particles are assigned a porosity \((\varepsilon)\) equal to the micro-porosity of the particles \((\varepsilon = \varepsilon_o)\), whereas for the cells outside the porous particles are assigned \(\varepsilon = 1\). The computational cells cut by the fluid/porous-medium interface, are assigned a porosity value in between \(\varepsilon_o\) and 1. As a result, one can correlated \(S\varepsilon\) and \(\varepsilon_o\) as, \(S(1-\varepsilon_o) = 1-\varepsilon\).

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The value of \(\beta\), for open-cell solid foams has been evaluated by means of fully resolved simulations in terms of a closure problem. Considering an idealized foam geometry (tetrakaidecahedron), a pressure drop or drag correlation has been derived using fully resolved

Fig. 2. A typical example for calculating \(\varepsilon\) and \(S\) when the porosity of the particles, \(\varepsilon_o = 0.9\). Computational cell A is completely inside a porous particle, whereas B is partially inside the particle, and C is a pure fluid cell.
pore-scale level simulation as reported by Das et al. [18]. The following form of it is used here:

$$\frac{\Delta p}{L} = 70.54 \frac{\mu(1-\varepsilon_p)^{1.38}}{d_p^{2.5}} (\overline{u}) + 1.95 \frac{\varepsilon(1-\varepsilon_p)^{1.18}}{d_p^{2.5}} (\overline{u})^2$$

(6)

The length scale, $d_p = 6V_{REV}/A_{tot}$ represents the equivalent spherical diameter of the porous foam, where $V_{REV}$ and $A_{tot}$ are the solid volume and surface area of the foam in the REV, $(\overline{u})$ represents the superficial velocity in the direction of the applied pressure gradient. For steady flow through a homogenous porous foam, in the absence of any wall effect, the momentum equation can be simplified as, $\varepsilon V(p)/ = \beta (\overline{u})'$. The momentum exchange coefficient $\beta$, in terms of interstitial velocity, $(\overline{u})' = \frac{1}{\varepsilon} (\overline{u})$, is given as,

$$\beta = 70.54 \frac{\mu(1-\varepsilon_p)^{1.38}}{d_p^{2.5}} + 1.95 \frac{\varepsilon(1-\varepsilon_p)^{1.18}}{d_p^{2.5}} (|\overline{u}|)$$

(7)

2.2. Solution methodology

In the current Finite Volume (FV) implementation the transport equations (Eqs. (4) and (5)) are integrated for each staggered computational control volume (CV) and the time-discretized form of the momentum equation is obtained as:

$$ρ\epsilon\left(\left(\frac{\partial}{\partial x}ight)^2\right)^{1.1} = ρ\epsilon\left(\left(\frac{\partial}{\partial x} \right)^2\right)^{1.1} \Delta V + Δε\frac{S(\Delta)}{\varrho}$$

(8)

where $V, A$ and $n$ represent the volume, face-area and face-area normal of the CV. The time level is indicated by the notation $k$. A deferred correction method is incorporated in the convection term (C = $ε V(p)/ + \n (\overline{u})'$), where both the First Order Upwind (FOU) scheme and the total variation diminishing (TVD) Min-mod scheme are used to calculate the convection flux. The flux based on the FOU scheme $(C^{1.1}_{FOU})$ is used as a predictor and treated implicitly, while the difference between the TVD and FOU schemes $(C^{1.1}_{TVD} - C^{1.1}_{FOU})$ is used as corrector that is treated in an explicit manner. A second order accurate Central Difference scheme is used for discretizing the diffusion terms ($F = μ V(p)/ + (\overline{u})'/\Delta V$). Eq. (8) is solved on a staggered computational grid by a fractional step method where, initially a tentative velocity field $((\overline{u})')$ is computed using the old time level pressure value $((\overline{u})')$: \n
$$\varphi (\overline{u})' = \varphi (\overline{u})' + \frac{Δt}{ΔV} \{ - \int_A (\varepsilon (\overline{u})' V) n dA \}$$

(9)

The stress tensor $(\tau)$ is split into an implicit part and an explicit part. In the momentum equation for the $x$ direction, only the $x$ velocity component is treated implicitly, whereas the other velocity components are treated explicitly. As a result Eq. (9) forms three decoupled sets of linear equations. To find $((\overline{u})')$ we use a robust and efficient parallel Block - Incomplete Cholesky Conjugate Gradient (B-ICCG) solver.

By subtracting Eq. (8) from Eq. (9) and taking the divergence, the Pressure Poisson Equation (PPE) is obtained as:

$$V \left( \frac{Δt}{\varrho \varepsilon \Delta V} - \varphi (\overline{u})' \right) = V.\left[ ε ((\overline{u})') \right]$$

(10)

where $\varphi (\overline{u})' = p^{k+1} - p^k$, represents the pressure correction. Again Eq. (10) is solved by using a B-ICCG solver. The velocity at the new time level $k + 1$ can be obtained from:

$$\Delta t = \frac{u_{m}}{2D_{a}}$$

(11)

It is important to mention here that the no-slip boundary condition at the cylindrical wall in a Cartesian domain is enforced by the immersed boundary method, detailed in our previous work [19].

2.3. Verification

In order to verify the present macroscopic model, a classical test case, Poiseuille flow through a 2D channel partially filled with a porous block is considered (Fig. 3-a). The porous medium is considered to be homogeneous, and inside the porous medium, creeping flow is assumed. The fluid enters the fluid/porous composite channel with an uniform velocity $u_0$ and the far-stream superficial velocity inside the porous medium is $U_0$. Flow inertia at both the fluid region and the porous medium are neglected, i.e. we consider axially fully developed flow.

For free fluid flows over permeable surfaces, Beavers and Joseph [28] proposed an empirical velocity-slip condition at the fluid/porous-medium interface by matching Darcy’s Law with the Navier-Stokes equations. They have suggested to use a uniform velocity profile (plug flow) in the porous medium (using Darcy’s law) and based on the permeability of the porous medium proposed an expression to calculate the slip velocity at the interface. As a result in their model, there exists a jump in the velocity profile. Neale and Nader [21] argued that, at the interface, the velocity of the fluid $(u_{fluid})$ should match the superficial velocity in the porous medium $(u_{porous})$. Furthermore, the macroscopic viscous shear stress at the interface is equal to the shear stress on the fluid side. Mathematically, the problem involves the coupling of the momentum equation for the fluid region (i.e. Navier-Stokes equation) with the equation for porous medium (i.e. the Brinkman-Darcy model) with the following matching conditions,
\[
\frac{dp}{dx} = \frac{d(p/\mu)}{dx} \bigg|_{\text{porous}} 
\]
(12)

\[
\langle u \rangle_{\text{porous}} \bigg|_{x=a} = U_0 = -\frac{K}{\mu} \frac{d(p/\mu)}{dx} \bigg|_{\text{porous}} 
\]
(13)

At the porous interface: \( u_{\text{fluid}} = \langle u \rangle_{\text{porous}} \)
(14)

\[
\mu \frac{d}{dx} (u_{\text{fluid}}) = \mu_{\text{eff}} \frac{d}{dx} (\langle u \rangle_{\text{porous}}) 
\]
(15)

We perform simulations for a 2–D channel, similar to the configuration shown in Fig. 3-a. The wall adjacent to the fluid region is subjected to the no-slip boundary condition, whereas the wall adjacent to the porous region is subjected to free-slip boundary condition. The present single domain formulation satisfies the conditions specified in Eqs. (12)–(15) inherently with \( \mu_{\text{eff}} = \mu \). The fully developed velocity profile calculated from the present CFD code is compared with the analytical velocity profile derived by Neale and Nader [21] in Fig. 3-b. Porous blocks of different porosity are simulated and an excellent agreement is obtained between the analytical and numerical results. The equivalent length scale of the porous medium (\( d_p \)) is considered as \( 15 \times 10^{-4} \text{ m} \) and viscosity, \( \mu = 0.1 \text{ Pa} \cdot \text{s} \). The flow is simulated at \( Re_0 = 1 \) (based on \( d_p \) and \( U_0 \)) and the permeability of the porous zone is calculated from Eq. (6). The height of the channel is \( 20 \times d_p \), large enough to omit the bottom free-slip wall effect on the velocity profile. It is clearly observed that the viscous shear in the fluid region at the porous interface, penetrates into the porous medium and forms a boundary layer region; the boundary layer thickness increases with increase in porosity (i.e. decrease in \( K \)).

3. Packing generation

There are two major approaches for generating a random packing structure: experimentally through tomography of the column [29] and numerically by the discrete element method (DEM) [30]. The DEM is typically used to simulate the flow dynamics of granular material where the motion of the particles is determined by applying Newton’s second law of motion to each particle. Each particle experiences a contact force when colliding with other particles or a wall and a body force due to gravity. Contact detection and contact force calculations for spherical particles are straightforward: only the center of mass locations of the particles are needed. However, for non-spherical particles, it is not trivial as the orientation of the particles also needs to be accounted for. Sometimes, it becomes very difficult to control the simulation for particles of high aspect ratio, where a small angular motion causes a larger linear deformation. Random packings of cubic and cuboid particles in the cylindrical columns are generated using a glued-sphere Discrete Element Method [31–33]. The glued-sphere approach overcomes all these difficulties and provides reasonable accuracy. In the glued-sphere approach, the outer shape of any non-spherical particle is represented by several overlapping small spheres (glued-spheres) where the relative motion of the glued-spheres in a particular non-spherical particle is restricted in the DEM simulation. The accuracy of the simulation increases with the number of the glued-spheres, however, it also increases the computational expenses. As sharp edges of the particles are not resolved, for highly dynamic granular flows it provides inaccurate and inconsistent results [31]. However, for the present cases i.e. generation of static packings of particles, it offers reasonably accurate results. We use the frame-work of an open-source DEM code LIGGGHTS [20] to generate the packings.

Once all the particle positions (center of gravity) and orientations (quaternions) are known, we require the calculation of the porosity field \( \varepsilon \) in the 3D Cartesian computational domain. At first, all the computational cells that are fully inside a porous particle are assigned \( \varepsilon = \varepsilon_p \) and pure fluid cells are assigned \( \varepsilon = 1 \). We call a computational cell fully inside the particle (fluid) when all the eight vertices of that cell are inside the particle (fluid). The surface of the particles are not necessarily aligned with the computational grids. The computational cells near the particle surfaces are partially occupied by the porous particles. In the present study, the porosity (3D polyhedral volumes formed by grid lines and particles) of such cells is calculated numerically, by dividing the computational cells into very small sub-cells [19]. By counting the number of sub-cells that are inside the particle, the particle volume in that cell and then porosity of that cell is calculated (detailed description in Das et al. [19]). In all calculations presented in this work, we divide each cell into \( 10 \times 10 \times 10 \) sub-cells, which provides us with an accuracy of ~0.1% with respect to the porosity calculation. A similar approach is used to calculate the porosity near the column wall, where cells are also cut by the cylindrical wall and hence decrease the cell volume. To minimize the computational expenses, at first, we loop over all the particles and create a bounding box around each particle. Subsequently, for each of the porous particles we only examine the computational cells inside the associated bounding box to assign its porosity value.

4. Results and discussions

Three different sized particles, cuboid and cubic shaped of dimensions \( 9 \text{ mm} \times 9 \text{ mm} \times 3 \text{ mm} \), \( 9 \text{ mm} \times 9 \text{ mm} \times 6 \text{ mm} \), \( 9 \text{ mm} \times 9 \text{ mm} \times 9 \text{ mm} \) are used to generate packings. These are denoted as P3,P6 and P9-particles, respectively. Random packings by using each of the particle types are generated in four different columns of diameter \( D = 30 \text{ mm} \), 55 mm, 83 mm and 120 mm. These are denoted as C30,C55,C83 and C120-column, respectively. This results in totally twelve different packing structures with different inter-particle voidage (\( \varepsilon_p \)). It should be noted that \( \varepsilon_p \) reduces to the overall porosity of the bed when the porosity of the particle, \( \varepsilon_p = 0 \). For most of the flow simulation cases, particles are considered to be composed of open-cell solid foam, with porosity \( \varepsilon_p = 0.9 \) and a specific surface area (surface area per unit volume of the foam sample) of \( S_f = 2600 \text{ m}^2/\text{m}^3 \). These values correspond to foams of equivalent spherical diameter \( d_p = 0.23 \text{ mm} \) (refer Eq. (6)). To study the effect of the particle porosity, for P9–C83 case, additional simulations are performed for two different particle porosities (\( \varepsilon_p = 0.80 \) and \( \varepsilon_p = 0.95 \)), while using the same \( d_p = 0.23 \text{ mm} \). The corresponding Darcy number (\( Da = K/d_p^2 \)) and non-dimensional Forchheimer coefficient (\( F_0 = (b/K)d_p \)), for the three different foam samples are tabulated in Table 1. From a hydrodynamic point of view, \( d_p \) represents the length scale for flow inside the porous particles. For flow outside the particles, i.e. through the inter-particle void spaces – following the packed bed literature – the length scale is defined as \( d_f = 6V_p/A_p \). Note that, \( V_p \) and \( A_p \) are the volume and outer surface area of the particles while the internal porosity of the particles is neglected. Hence, \( d_p \) represents the equivalent spherical diameter of the non-spherical particles. For P3,P6 and P9 particles, we obtain \( d_f = 5.4 \text{ mm} , 7.714 \text{ mm} \) and 9 mm, respectively.

Similar to the two different length scales, i.e. \( d_p \) and \( d_f \), we define two different velocity scales \( U_0 \) and \( U_f \) for flow through and around the porous particles. The velocity \( U_t \) is the inlet superficial velocity of the full column, whereas \( U_f \) represents the average superficial velocity inside the particles, calculated numerically as a post-processing step. Flow simulations are performed for the C30,C55 and C83 column where the macroscopic Reynolds number (\( Re_{d_f} \) – based on \( d_f \) and \( U_f \) – is varied from 0.01 to 400. To characterize the flow inside the particles, we

<table>
<thead>
<tr>
<th>( \varepsilon_p )</th>
<th>( d_p (\times 10^{-3} \text{ m}) )</th>
<th>( S_f (\text{m}^2/\text{m}^3) )</th>
<th>( Da = 1 \times d_p^2/N )</th>
<th>( F_0 = \frac{b d_f}{K} )</th>
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Table 1

The properties of different foam samples used in the present study.
define a microscopic Reynolds number \((Re_d)\) based on \(d_c\) and \(U_c\). The DEM simulation time for the C120–P3 packing (2100 cuboid particles with 602 glued-spheres per cuboid particles) is approximately 7 days in a single core of Intel Xeon 2.60 GHz processor. For hydrodynamics simulation of the C83–P3 case (~ 45 million mesh), it requires approximately 5 days of simulation time using 24 cores (Intel Xeon 2.60 GHz processor) to reach steady state (up to ~ 1000 time-steps).

4.1. Porosity and packing structures

In general, the bed porosity depends on (i) particle to column diameter ratio, (ii) particle size and shape, (iii) mechanical properties of the particles, (iv) loading method, and (v) the external agitation of the column or fluidization. There is no generalized correlation which relates the bed porosity with all these parameters, not even for spherical packings [34]. At the same time, for non-spherical particles, the initial location and orientation of the particles during the filling process change the bed porosity drastically. Initially, clusters of non-spherical particles are randomly placed near the top of the column and allowed to fall due to gravity. In such a single cluster, the particles are placed randomly and oriented arbitrarily by a Monte Carlo simulation procedure available in LIGGGHTS. Several groups or clusters of particles are used to fill the column up to the desired height of approximately \(H = 15d\). After insertion of the final group of particles, the DEM simulation is continued until all particles have come to rest. The DEM simulation parameters are listed in Table 2. A maximum particle overlap of \(1\%\) of the glued-sphere diameter is obtained from the DEM code. We use particles of a specific roughness (coefficient of friction) without any external agitation of the column. In this case, the packing generation is random due to the arbitrary placement and orientation of the groups of particles; and the number of particles per group.

We first study the effect of initial random placement of particles on the resulting packing porosity by running the same simulation several times (with different seeds for random number generator) while keeping all other settings the same. We found a maximum deviation of \(3\%\) on the overall porosity. Using the Ergun correlation, one can estimate \(\sim 10\%\) of the variation in pressure drop due to the randomness of the packing structures. However, in this study, we restrict ourselves to a single packing structure for a specific column-particle combination. Subsequently we perform three different simulations only varying the number of particles per group such that the porosity of the initial groups becomes approximately 0.7, 0.8 and 0.9, and here too, we found negligible differences on the overall porosity. However, if we change the mechanical properties of the particle, it may change the porosity of the bed. For the present filling method and the particle properties, we can conclude that the packing structure is more or less the same and that it is reproducible. The final packing structures for P3 and P9 particles for different column diameter are shown in Fig. 4. We will now present a quantitative analysis of the different packing features in the subsequent subsections. In this analysis we consider particles that are non-porous or solid, i.e. \(\eta = 0\).

4.1.1. Particle distribution

Fig. 5 shows the distribution of the particle center of gravity (CG) projected on the cross-section of the cylinder for different packing structures. Though the particles are randomly packed in the column, near the wall they have an ordered packing due to the presence of the wall, regardless the particle shape and column diameter. First, let us consider the packings of P9 (cubic) particles in the different columns. Near the wall, a sharp ring-like structure of particle clusters are formed due to the ordered particle alignment. The ring-like structure propagates towards the center of the column, at the same time becoming more difuse. At larger column diameters, the ring-like structure becomes more apparent and can be observed at a greater distance from the column wall. The average distance between the rings is the equivalent diameter of the P9 particle. On the contrary, for P3 particles, near the wall two distinct rings are observed for all the columns. Due to the different side-length, P3 particles have two options to orient near the wall: either the smaller face touches the wall or the larger face is oriented towards the wall. In these two different configurations, two different particle clusters form due to a different wall-to-particle (CG) distance. Particles have a natural tendency to align horizontally (minimum potential energy state) and, as a result, the outer ring is less dense compared to the inner ring. Both rings prorate inward and interfere with each other, eventually producing a homogeneous particle clustering at the core of the column just one particle diameter away from the wall.

4.1.2. Particle orientation

The angle between the axis of a particle and the column axis (represented by \(\theta\)) is calculated for all the particles and the Number Density Function (NDF) of the orientation of the particles \(\theta\) for different packing structures are shown in Fig. 6. Fig. 6-(a to c) shows the NDF for the P3 particles where for all the column diameters, the NDF increases monotonically with \(\theta\). This indicates that the particles tend to align orthogonally to the column axis (minimum potential energy state). This alignment pattern becomes more pronounced with increasing the column diameter. The orientation distribution for P6 particles is shown in Fig. 6-(d to f). Although in this case too the particles are more prone to align horizontally, but as the aspect ratio of P6 particles is comparatively smaller, fluctuations are observed in the NDF plot. The magnitude of this fluctuation decreases with increase in column diameter. Finally, Fig. 6-(g to i) shows the orientation NDF for P9 particles. It should be noted that as P9 particles are cubic shaped, both \(\theta = 90^\circ\) and \(\theta = 0^\circ\) describe a similar particle orientation. As a result, NDF plots are symmetric along \(\theta = 45^\circ\). Also, for large column diameters the NDF profiles are almost flat for P9 particles, due to their cubic shape. However, for C55 and C88 columns, there are peaks at \(\theta = 0^\circ(90^\circ)\), which is due to bottom wall effects, causing the particles to be aligned perfectly, primarily in the bottom section of the column (please refer to Fig. 4).

4.1.3. Axial and radial porosity profile

For a very large column, the global porosity is generally calculated on basis of the number of particles and the packing height. For a small column, the ordered packing structure near the bottom wall may contaminate the result. This effect can be observed in Fig. 7 which shows the axial porosity \((\eta_a)\) profile along the normalized axial distance \(m = x/d_i\). Here, \(x\) and \(d_i\) are the axial coordinate system and smallest edge length of the particles, respectively. The fluctuation in the axial porosity is more pronounced for smaller column diameter to particle

<table>
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<th>Parameters</th>
<th>Value</th>
<th>Units</th>
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<td>P9</td>
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<td>Equivalent diameter ((d_C))</td>
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equivalent diameter ratio (Fig. 7-a,b). For large column diameters the amplitude and frequency of this fluctuation decreases, indicating a closed packing structure. In the bottom of the C120 column (Fig. 7-c,d) high fluctuation in $\varepsilon_a$ is found for $(m = 0 - 3)$ indicating bottom wall effects on the structure of the packing. Here, we are interested in a packing structure which can be applied for very tall columns. Hence, we only use the middle section of the generated packings from DEM simulations as an input for the CFD domain, and also for the calculation of bulk porosities ($\varepsilon_p$) and radial porosity profiles ($\varepsilon_r$). By doing so, we have a more or less homogeneous axial bed porosity throughout the bed free from inlet (bottom) effects.

The radial porosity profile is an important input parameter to quantify any wall channeling effect. Moreover, knowledge of the radial porosity profile is important to homogenize the full bed for axisymmetric coarse-grained models. Fig. 8 shows the radial porosity profile ($\varepsilon_r$) versus non-dimensional wall distance ($p = (R - r)/d$). Here, $R = 0.5D$ is the radius of the column. Recall that in this analysis we consider $\varepsilon_r = 0$. All the plots start from $\varepsilon_r = 1$ at the wall, showing an oscillation in the porosity near the wall that damps out towards the center of the column (except for the very slender C30 column). The particles near the wall have an ordered packing. The oscillation in $\varepsilon_r$ near the wall is due to the alignment of the outer ring of the particles against the wall, whereas in the core section, the particles are oriented very randomly, creating a homogeneous porosity distribution.

For the largest column diameter, i.e. for C120, the range of this oscillation extends to $p \sim 3$ and decreases with a decrease in column diameter. The amplitude of the fluctuations also decreases from P9 to P3 particles, i.e. with a decrease in the size of the particles, as smaller particles can very closely align against the wall. For the P9 particles, sharp peaks in $\varepsilon_r$ are observed at the integer intervals of the non-dimensional radial position $p$, i.e. at $p = 1, 2, 3, 4$ etc. Very similar behavior can also be observed for the packings of spherical particles. However, in that case the oscillations of $\varepsilon_r$ are more smooth, and peaks are observed at $p = 1, 2, 3, 4$ etc., whereas valleys are observed at $p = 0.5, 1.5, 2.5, 3.5$ etc. [19]. For spherical packings, $\varepsilon_r$ is high at the center of the layer of spherical particles touching the wall, and low at the location where two layers (of spherical particles) touch each other. In contrast, cubic particles (P9) have a higher tendency to contact the wall by face. Also, two layers of particles mostly touch each other by face and very few by corner points or by face. As a result, $\varepsilon_r$ changes sharply from low to high at $p = 1, 2, 3, 4$ etc. For P6 and P3 particles, the situation is even more complicated. Near the wall, fluctuations of different amplitude and frequency can be seen. As mentioned earlier, P6 and P3 particles have mainly two tendencies to align with the wall, either the smallest face touches the wall or the largest face is directed towards the wall. Few particles touch the wall by their edge or corner points. The different amplitude in near wall fluctuation of $\varepsilon_r$ can be attributed to the different tendency of particles to align vertically or horizontally against the wall. For P3 and P6 particles, for the same column to particle diameter ratio, the span of the near wall region is less compared to the P9 and spherical particles.

4.1.4. Overall bulk porosity (inter-particle voidage)

The bulk porosity ($\varepsilon_p$) is an important parameter to characterize the
full bed for engineering applications. The pressure drop and the overall bed-to-wall heat transfer are a strong function of the bulk porosity. Using the current DEM simulation data, we examine the effect of the particle aspect ratio and column to particle equivalent diameter ($N = D/d_p$) on the overall bed porosity.

For structured spherical packings, the lowest porosity $\varepsilon \approx 0.26$ can be obtained for face-centered cubic (FCC) configuration. However, one can achieve $\varepsilon \approx 0$ by simple cubic (SC) arrangement of cubic or cuboid particles. This simple example indicates that a larger range of overall porosity can be obtained by packings of non-spherical particles. For random packings, as expected, the bed porosity decreases with increase in $N$. For packed beds with spherical particles, the overall bed porosity is a smooth function of $N$ and reaches a constant value of $\varepsilon = 0.37$ for very large column diameters ($N \to \infty$). This is not the case for non-spherical particles [30,29]. In random non-spherical packings, the particle aspect ratio and the orientation significantly change the overall bed porosity and as a result, the (transport) performance of the bed. For the present case, the overall porosity for different packing structures is listed in Table 3. The bulk porosity varies from 0.245 to 0.475, which is significantly broader than the range observed for spherical particles. The general observation is that for the same column an increasing particle size leads to an increase of the bed porosity. Similarly, for the same particle size, an increase in column diameter leads to a decreased bed porosity. In Fig. 9 the variation of $\varepsilon$ with $N = D/d_p$ for different packing structures is plotted. It shows a smooth decreasing trend of $\varepsilon$ with $N$ for packings of P9 and P6 particles, however for P3 particles this variation is not smooth due to the higher aspect ratio.

4.2. Hydrodynamics

The primary objective of the flow simulation is to compute the pressure drop and velocity distribution for different packing structures. To remove boundary effects, for all the simulations, an inlet section and an outlet section are provided at the upstream and at the downstream of the packing structure. At the inlet, a uniform plug flow velocity of $U_s$ is specified. The height of the packings is almost $10 \times d_p$ for all cases, in order to allow the flow to become fully developed. Fig. 10 shows the simulation snapshots for a typical case (C83–P9,$Re_d = 25$ and $\varepsilon = 0.9$). Contours of the axial component of the interstitial velocity $(\langle u_z \rangle)$ normalized by the superficial velocity of the bed ($U_s$) are shown at a particular longitudinal and a transverse section of the bed. In the present study, we also consider the presence of the cylindrical wall. With
Fig. 6. Number density function (NDF) of the global orientation of the particles. $\theta$ represents the angle between the axis of a particle with the column axis.

Fig. 7. Averaged axial porosity profile ($\varepsilon_a$) with normalized axial distance ($m$) for different packing structures. Only the middle section of the packings is used in CFD simulations and to calculate the average bed porosity. It should be noted that in this calculation the porosity inside the particles are not considered, i.e. $\varepsilon_w = 0$. 

$S.\text{Das et al., Chemical Engineering Journal 334 (2018) 741–759}$
the present numerical model, it is possible to compute the distribution of the flow through and around the porous particle. From the streamlines plot the flow around and through the particles are clearly observed. The fluid experiences different resistances while flowing through and around the particles. Velocity is high in the interstitial spaces between the particle as compared to the velocity inside the internal pores of the particles. The internal pores allow the fluid to flow through the particles. As a result it reduces the tortuosity and the

Fig. 8. Averaged radial porosity profile (εr) with normalized wall distance (p) for different packing structures. In this calculation porosity inside the particles are not considered, i.e. εi = 0.
associated pressure drop, compared to the same packings of solid particles.

As the flow through the interstitial spaces between the particles is resolved, a proper grid convergence study is performed for a typical case (see Appendix A). Cartesian grids of equal spacing ($\Delta x = \Delta y = \Delta z$) are used and our grid convergence study suggested that 20 grid cells across the shortest length of the particles provide grid independent results. Consequently we chose $\Delta x = \Delta y = \Delta z \sim 3 \times 10^{-4} m$. Depending on packing structures, the total number of computational cells vary from $\sim 6 \times 10^6$ to $\sim 45 \times 10^6$. The time-step, $\Delta t \sim 10^{-5}$ is chosen to obey the Courant–Friedrichs–Lewy (CFL) criterion for all the cases.

### 4.2.1. Pressure drop

For flow through homogeneous porous media, at very low fluid velocity, the pressure drop is only balanced by the viscous shear stress at the solid surfaces (Darcy regime). When the Reynolds number is gradually increased, in the Forchheimer regime, the inertial forces related to the local acceleration of the fluid particles start to contribute and the total pressure drop contains both viscous and inertial contributions. The exact limiting value of the Reynolds number for flow transition from Darcy regime to Forchheimer regime depends on the porosity of the porous medium, as-well-as the definition of the length scale to calculate the Reynolds number.

The pressure drop due to viscous stress is proportional to the superficial velocity ($U_s$) whereas the inertial contribution in the overall pressure drop in porous media is expressed by a $U_s^2$ term by almost all authors, and can be justified by the time-integration of the average D macroscopic advection term ($\langle u_c \rangle \cdot V_u$). The most classical and widely

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<th>$C_83$</th>
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</table>

**Table 3**

Bulk porosity ($\epsilon_P$) of the bed for different particle column combinations; it should be noted that in this calculation the porosity inside the particles is not considered, i.e. $\epsilon_i = 0$.

Fig. 9. Overall bed porosity ($\epsilon_P$) with column to particle equivalent diameter ratio for three different types of particles. It should be noted that in this calculation the porosity inside the particles is not considered, i.e. $\epsilon_i = 0$. 

Fig. 10. Simulation snapshots for $C83$–$P9$ case operating at $Re_d = 25$ with $\epsilon_0 = 0.9$. Contours of normalized interstitial axial velocity ($\langle u_x \rangle / U_s$) at a particular longitudinal and transverse section of the bed are shown. The streamlines clearly exhibit the flow around and through the particles. Velocity is high at the interstitial spaces between the particle compared to velocity inside the internal pores of the particles. In a Cartesian grid the no-slip condition at the column wall has been imposed by an immersed boundary method.
used relation for predicting pressure drop over a packed bed with spherical and non-spherical solid particles has been suggested by Ergun and reads:

$$\frac{\Delta p}{\Delta x} = \alpha \frac{\mu}{\phi} d_{eq}^2 U_i + \beta \frac{\rho}{\phi} d_{eq} U_i^2$$

(16)

The values of $\alpha$ and $\beta$ depend on the type and the structure of the porous medium. For packed beds filled with spherical or non-spherical particles, Ergun proposed:

$$\alpha = A(1-\varepsilon_p)\frac{A}{d^2_{eq}}, \beta = B(1-\varepsilon_p)\frac{A}{d^2_{eq}}$$

and $A = 150, B = 1.75$

(17)

The length-scale $d_{eq}$ represents the diameter of the equivalent-volume sphere and the shape factor $\phi$ is the sphericity of the particles, which is defined as the ratio of the surface area of equivalent-volume sphere ($A_{eq}$) to the surface area of an actual particle ($A_p$):

$$\phi = \frac{A_{eq}}{A_p} = \frac{1}{\pi}(6V_p)^{\frac{1}{3}}$$

(18)

where $V_p$ is the volume of the particle. For a bed packed with spherical particles, the sphericity $\phi$ is taken as unity and the diameter $d_{eq}$ is the actual diameter of the particles. As mentioned earlier, we chose the equivalent spherical diameter of the particles, $d_f = 6V_f/A_p$ as a characteristic length scale and these two diameters correlate as,

$$d_{eq} = \frac{6V_f}{A_{eq}} = \frac{6V_f}{A_p} = d_f \phi$$

(19)

With the use of the current definition of the equivalent diameter $d_f$, representing the diameter of spheres that would have the same volume to surface area ratio, the shape factor disappears in Eq. (16). For the present P3, P6 and P9 particles the values of $d_f$ are 5.4, 7.71 & 9 mm and $\phi = 0.679, 0.791 & 0.806$. Although Ergun proposed universal values of the constants $A$ and $B$ in Eq. (17) for viscous and inertial terms, both may vary based on the particle aspect ratio and shape [10,35,11,36], and the slenderness of the column [7,8,19]. It is well-known that the Ergun constants fit the experimental/simulation data of spherical particles packed in a large column well and reasonably accurate (20–30%) for non-spherical particles. Several studies are focused on correlating the effect of the column to particle diameter ratio on the pressure drop for mono-disperse spherical packings. However, to the best of our knowledge, very little is available in literature considering the effect of the column diameter on the pressure drop for non-spherical particles. In almost all studies for spherical packings, the porosity function, i.e. $(1-\varepsilon_p)^2/\varepsilon_p$ for the viscous term and $(1-\varepsilon_p)/\varepsilon_p$ for the inertial term first proposed by Ergun were found to be very accurate [7,8,19]. Hence, the focus was to find the values of $A$ and $B$ for dense packings. For non-spherical particles the situation is even more difficult, as it demands a proper choice of the equivalent diameter/length scale that obeys an Ergun type porosity function for all particle shapes and aspect ratios. For flow through packings of ellipsoids, Rong et al. [11] used $d_{eq}$ as the characteristic diameter, whereas Dorai et al. [10] used $d_f$ as the characteristic diameter for flow past packings of cylindrical particles. In both the studies wall effects are neglected. Experimental work by Li et al. [35] suggests using $\phi \times d_f$ as the equivalent diameter. The main motivation behind the choice of different length scales is to make $A$ and $B$ independent of porosity. In the present case, we use $d_f$ as a length scale and $U_i$ as a velocity scale for the full bed. To compare the present results the classical form of the Ergun correlation is used.

In this work, we normalize the pressure gradient using the viscous scale and hence the non-dimensional form of Eq. (16) reads:

$$f = \frac{\Delta p}{\Delta x} \epsilon \frac{\mu U_i}{d_f^2} = \alpha + \beta Re_{dp}$$

(20)

As a result, for fluid flow through the interstitial spaces between the solid particles of a packed bed, the non-dimensional pressure drop ($f$) becomes a linear function of the Reynolds number ($Re_{dp}$). In contrast, for the present situation at hand, the fluid can take two possible pathways, either through or around the particles. Hence, it is not expected that $f$ will depend linearly on $Re_{dp}$. The Reynolds number ($Re_{dp}$) constitutes the superficial velocity ($U_i$) and the equivalent spherical diameter ($d_f$), describing the flow outside the particles. However, to characterize the flow inside the particles, we define $Re_{dp}$ based on the length scale of the open-cell solid foam ($d_f$) and the superficial velocity inside the foam particles ($U_f$) (refer Eq. (21)). The distribution of flow inside the particles (micro-pores) and in the interstitial space outside the particles (macro-pores) depends on the differences in flow resistance. Based on the relative contribution of the viscous force and flow inertia, occurrence of three different flow regimes can be identified: (a) creeping or Stokes flow at both the micro and macro-pores, i.e. $Re_{dp} \to 0$ and $Re_{dp} \to 0 (b)$ creeping flow at the micro-pores, but inertia dominated Forchheimer flow in the macro-pores, i.e. $Re_{dp} \to 0$ but $Re_{dp}$
is finite; and (c) Forchheimer flow at both the micro and macro-pores, i.e. both $Re_\mu$ and $Re_{dp}$ are finite.

Fig. 11-(a) shows the inverse of the permeability, $K^{-1}$ with inter-particle voidage ($\epsilon_p$) at $Re_{dp} = 0.1$ ($Re_\mu \approx 10^{-3}$) of foam particles with $\epsilon_p = 0.9$ and $d_p = 0.23$ mm. For such low inertial flow the pressure drop is directly proportional to the available solid surface area. For the C83 column with different particles, $K^{-1}$ i.e. flow resistance decreases almost linearly with increase in bed porosity. However, for the C55 and C30 columns $K^{-1}$ versus $\epsilon_p$ show stiff and/or variable slopes. This behaviour reflects the combined effects of the cylindrical wall, particle shape factor and inhomogeneity in the porosity distribution (i.e. presence of $\epsilon_p$ and $\epsilon_\mu$ simultaneously.).
pressure drop cannot be captured by Kozeny–Carman [37] type equations. For the nine cases studied in this work, we cannot be fully conclusive on this variation. The relative contribution of the two different flow resistances (due to $\varphi_p$ and $\varphi_s$) governs the flow-rate inside and outside the porous particles and as a result the overall pressure drop.

The subsequent analysis focuses on the pressure gradient when flow inertia starts to build up. Fig. 12 shows the variation of $f$ with $Re_d$ for different packing structures. The macroscopic Reynolds number of the flow is varied up to $Re_d = 400$. Due to the very small length scale at the micro-pores, the Reynolds number inside the particles, $Re_p$, is less than 14, for all the cases. We define the limit of the Darcy region inside the particle up-to $Re_p < 1$, when the relative contribution of the inertial terms to the pressure drop is less than 4% (Eq. (6)). In Fig. 12 the open squares ($\square$) indicate the transition of flow from Darcy to Forchheimer regime inside the particles. Up to approximately $Re_p = 1$, the overall non-dimensional pressure drop of the bed varies linearly with the macroscopic Reynolds number, i.e. $f = \alpha + \beta Re_d^{0.68}$. This is attributed by a constant non-dimensional pressure drop or drag inside the porous particles (due to creeping flow) while fluid inertia is predominant for flow outside the particles. The slope of $f$ versus $Re_d$ decreases when flow inertia inside the particles gradually increases and it follows $f = \alpha + \beta Re_d^{0.68}$. Compared to beds with solid particles where $f \sim Re_d$, particles with internal pores scaling like $f \sim Re_d^{0.68}$ have an additional advantage in pressure drop reduction for increasing flow Reynolds numbers. For all the cases, irrespective of the bed porosity, the effect of the confining wall and the particle shape, similar $f \sim Re_d^{0.68}$ trends are observed. This indicates that the power $0.68$ is governed by the properties of the foam. In Table 5, the constants ($\alpha$ and $\beta$) are listed for all fits presented in Fig. 12.

The further analysis is dedicated to the effect of internal porosity ($\varphi_i$) of the particles on the overall pressure drop. Simulations are performed for the C83–P9 packing structures where the internal porosity of the particles is changed. Two different types of foam samples with porosity $\varphi_i = 0.8$ and $\varphi_i = 0.95$ ($d_p = 0.23$ mm) are used (Table 1) and similar to other simulation cases the macroscopic Reynolds number of the flow is varied up to $Re_d = 400$. Fig. 13 shows the variation of $f$ with $Re_d$ for different internal porosity of the particles. Increase in $\varphi_i$ indicates a smaller resistance to fluid flow inside it. Hence, the overall pressure drop decreases from $\varphi_i = 0.80$ to $\varphi_i = 0.95$. At the creeping flow condition, this can be analyzed simply by comparing the non-dimensional permeability or Darcy number of the different foam samples (Table 1). However, at the Forchheimer regime the non-linear inertial term changes the form of $\partial f / \partial Re_d$. For $\varphi_i = 0.90$, the curve of $f$ versus $Re_d$ exhibits a $f \sim Re_d^{0.68}$ trend, whereas for $\varphi_i = 0.8$ and $\varphi_i = 0.95$ it leads to $f \sim Re_d^{0.70}$ and $f \sim Re_d^{0.65}$, respectively. By increasing the porosity of the particles the Reynolds power also increases, albeit very slowly. It also indicates that only the power of $Re_d$, is a function of the internal porosity and length scale of the particles. The two non-linear terms in the pressure drop and prevailing flow distribution inside and outside the particles are the reason behind the present variation of $f$ with $Re_d$.

4.2.2. Flow division

The transport characteristics of packed bed reactors and the bed-to-fluid heat transfer depend on the rate of perfusion [15,16] which is directly related with average fluid velocity inside the particles. To quantify the average amount of fluid flowing through the particle, we define a flow distribution factor ($\Gamma$), which is the ratio of the average superficial velocity inside the particles ($U_i$) to the superficial velocity of the full bed ($\Gamma_i$):

$$U_i = \frac{\int_{V_{bed}} S (u_x) dV}{\int_{V_{bed}} S dV} ; \quad \Gamma = \frac{U_i}{U_i}$$

(21)

where $S$ is the indicator function (refer Fig. 2) and $<u_x>$ the superficial velocity in the axial direction. $V_{bed}$ represents the total volume of
curves (Fig. 11-(a)). The variation of 0 region is higher for (i) compared to:

\[
\frac{\partial}{\partial \Gamma} C_\text{p} \text{ versus } P_\text{fl}
\]

It should be noted that to calculate foam samples and decreases with 4

\[
\frac{\partial}{\partial \Gamma} C_\text{p} \text{ versus } P_\text{fl}
\]

decreases almost linearly with increase in P, and as a result the

total volume of the particles. For the C83 column filled with different

colour with porous particles (\( \partial \Gamma \)). For a packed bed with solid particles (i.e. \( \varepsilon_p = 0 \)) \( \Gamma = 0 \) and when there is no interstitial space between the porous particles (i.e. \( \varepsilon_p = 0 \)) \( \Gamma = 1 \). Apart from these two extreme cases, higher values of \( \Gamma \) indicate that flow has a higher tendency to penetrate the porous particles.

Fig. 14 shows the flow distribution factor (\( \Gamma \)) for different packing structures at creeping flow. For the C83 column filled with different sized particles, \( \Gamma \) decreases almost linearly with increase in \( \varepsilon_p \), i.e. with decrease in the total volume of the particles. For the C55 and C30 columns, \( \Gamma \) decreases too with increase in \( \varepsilon_p \), however, it exhibits a different slope. The behavior of the \( \Gamma \) versus \( \varepsilon_p \) curves is very similar to the \( K^{-1} \) versus \( \varepsilon_p \) curves (Fig. 11-(a)). The variation of \( \Gamma \) with \( \varepsilon_p \) can be explained, qualitatively, by comparing the flow resistance through a continuous foam sample with a packed bed containing solid particles.

For C83–P3, C83–P6 and C83–P9 packing structures, the ratios of these two flow resistances are calculated as 0.51 2.49 and 4.12, respectively. It indicates that the intra-particle flow resistance is maximum for C83–P9 packings, and as a result the fluid has a higher affinity to flow around the particles, i.e. a lower value of \( \Gamma \). This analysis clearly indicates the behavior of \( \Gamma \) versus the \( \varepsilon_p \) curve. Note that this can only be done for a larger column where the wall effects are minimal. To predict the flow resistance or permeability for flow through the interstitial spaces between the particles, the Ergun correlation is used, which is reasonably accurate for non-spherical particles at creeping flow conditions, whereas, to calculate the flow resistance for a continuous foam sample Eq. (6) is used.

In Fig. 15, the variation of \( \Gamma \) with \( \Delta R_\text{DP} \) is shown for different cases. For Darcy flow inside the particles, \( \Gamma \) increases linearly with \( \Delta R_\text{DP} \) and \( \delta \Gamma/\delta \Delta R_\text{DP} \) is slightly higher for P3 particles in different columns. A higher value of \( \delta \Gamma/\delta \Delta R_\text{DP} \) is also desirable as it will generally improve heat transfer performance and species conversion rates (in case of fast catalytic reactions). With increasing macroscopic Reynolds number \( \delta \Gamma/\delta \Delta R_\text{DP} \) decreases due to the increase of the inertial flow resistance inside the porous particles.

It is expected that when the viscous drag and the form drag resistance of the porous foam particles is decreased, more fluid will pass through the particles. The effect of \( \varepsilon_p \) on \( \Gamma \) is shown in Fig. 16 for the C83–P9 packing structure, which shows the expected behavior. Also it is interesting to observe that the slope of the curves, i.e. \( \delta \Gamma/\delta \Delta R_\text{DP} \) at the low \( \Delta R_\text{DP} \) region is higher for \( \varepsilon_p = 0.95 \) foam samples and decreases with \( \varepsilon_p \).

4.2.3. Drag reduction

The use of porous particles instead of solid particles increases the total available active surface area of the bed for both heat and mass transfer, and at the same time reduces pressure drop due to increase in the overall porosity (combining both \( \varepsilon_{\text{cell}} \) and \( \varepsilon_{\text{p}} \)) of the bed. Apart from the available surface area, the fluid-solid heat or mass transfer coefficients also depend on the local velocity of the fluid in contact with the solid surface. Very crudely, for the same interstitial velocity of fluid at inter- and intra-particle spaces, the total fluid drag reduction can be calculated in two ways. One can compare the pressure drop of the bed when it is filled with porous particles with the same bed containing solid particles when the average interstitial velocity (i.e. \( U_{\text{fl}}/\varepsilon_{p} \)) at the inter-particle space is the same. Another way is to compare the pressure drop in the bed with a continuous foam sample (similar \( \varepsilon_{p} \) and \( d_p \)) for the same average interstitial velocity (\( U_{\text{fl}}/\varepsilon_{p} \)). In Fig. 17, \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{C83}} \) represents the calculated pressure gradient for the C83 columns, \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{C83}} \) represents the pressure gradient when the same column would be filled with solid particles having the same inter-particle interstitial velocity (i.e. \( U_{\text{fl}}/\varepsilon_{p} \)) and \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{Foam}} \) corresponds to the pressure gradient of a continuous open-cell foam block with the same intra-particle interstitial velocity (\( U_{\text{fl}}/\varepsilon_{p} \)). It should be noted that to calculate \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{Foam}} \), the classical version of the Ergun equation (Eq. (16) and (17)) is used whereas \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{C83}} \) is calculated using the drag closure for open-cell foam (Eq. (6)).

The ratio of \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{Foam}} \) to \( \frac{\Delta \Gamma}{\Delta \varepsilon_{p}} \bigg|_{\text{C83}} \) with varying Reynolds number is shown in sub-figure (a). All values are below unity, which indicates the reduction in fluid drag for the case of a bi-disperse porous medium for a similar interstitial velocity. When a fluid flows over a porous particle, it penetrates into the porous layer (Fig. 3-(a)). As a result, from a macroscopic point of view, the surface of the porous particle exhibits a partial-slip boundary condition, whereas for solid particles a no-slip boundary condition applies. This is the reason for reduction in pressure drop for a bed with porous particles. For the same reason, in Fig. 16-(b)
the ratio $p_{C_{30}}$ to $p_{Foam}$ is less than unity. Except the lower range of Reynolds number, the ratio $p_{C_{30}}$ to $p_{Foam}$ is almost constant with $Re_{dp}$. With increasing macroscopic porosity ($ε_P$), $p_{C_{30}}$ and $p_{Foam}$ increases, whereas $p_{C_{83}}$ and $p_{Foam}$ decreases.

4.2.4. Radial velocity profile

Knowledge of the radial velocity profile ($U_r$) is required for further studies of the conversion rate and process evaluation in terms of the wall-to-bed heat transfer and undesirable flow channeling near the wall. The azimuthal-averaged interstitial axial velocity ($\langle u_x \rangle$) at each cross-section of the column is averaged along the length of the column to calculate the radial velocity profile. In Fig. 18, the radial velocity profiles ($U_r$) normalised by the superficial velocity ($U_s$) of the bed are plotted against normalized radial distance ($p$) for different cases. It can clearly be seen that the velocity profile follows the trend of the radial porosity profile (Fig. 8), and as expected at locations of high porosity, high velocities are observed. Near the wall, a very sharp gradient of the velocity profile is formed to facilitate the no-slip boundary condition at the wall. For the $C30$–$P3$ column, wall channeling is clearly visible up to $p = 1$ and $p = 2$, respectively. The peak velocity near the wall for $C30$–$P3$ column is almost 150% higher than the average velocity in the core, and for $C83$–$P3$ column it is around 75% higher. Sub-figures (a) to (c) also reveal the relative difference in velocity profile for high and low Reynolds number flows. With $Re_{dp}$, the increase in flow rate through the micro-pores of the particles, (i.e. increase in $Γ$) elevates the total interstitial velocity of the bed. As a result, the average velocity is slightly higher (at least in the core region of the bed) for $Re_{400}$ flows as compared to creeping flows. Finally, sub-figure (d) shows the effect of the internal porosity of the particles, i.e. $ε_μ$ on velocity profile for $C83$–$P9$ column. The peak velocity and the amplitude of fluctuation decreases with increases in $ε_μ$, i.e. internal flow resistance in the micro-pores. The fluctuation in the velocity profile is mainly caused by the inter-particle spaces (i.e. $ε_π$), as the velocity field is almost homogeneous inside the particles. With increasing $ε_μ$, the flow gets pushed more towards the intra-particle micro pores, which leads to lower velocity fluctuations.

4.2.5. Local velocity distribution

A powerful way to visualize and study a 3D velocity field in a complex porous medium is to analyze the probability density ($PD$) of the normalized interstitial velocity ($⟨u_x⟩/U_s$). By plotting the $PD$, one can study the variance or spread of the velocity field, and also identify
the degree of recirculation. At first, we focus on analyzing the PD of the velocity field for the intra-particle micro-pores. Fig. 19-(a) shows the velocity distribution inside the particles for the CP83–P9 column with ε0 = 0.9 in a linear scale. A very small recirculation zone can be identified by observing the negative tails in the PD plots. The size of the recirculation zone is very small and only observed near the contact point of the sharp edges of the particles. For all the cases a sharp peak and a long positive tail are observed. The spread or the variance of the intra-particle velocity increases with Reynolds number. With increasing \( Re_{dp} \), the boundary layer formation at the fluid-porous interface becomes more pronounced and a thick boundary layer inside the porous particle increases the spreading of the velocity variation. The shifting of the peaks of the PD curves towards the right shows the increase in mean velocity with increasing Reynolds number. The same plots on a logarithmic scale (Fig. 19-(b)) help us to identify the span of positive and negative tails. Here too, the number of recirculation cells (negative velocities) is very small.

5. Conclusions

We have investigated the flow behaviour through cylindrical fixed-bed reactors with randomly packed porous non-spherical particles. For
the present bi-disperse porous medium \((\epsilon_c \text{ and } \epsilon_s)\), a multiscale modeling approach, derived using volume averaging theory (VAT) has been developed. The flow at a length scale inside the particles i.e. \(d_p\) is not fully resolved, and represented using closure terms, derived separately from fully resolved computations [18]. However, the inter-particle flow (at the length scale of \(d_p\)) is fully resolved. A verification test case is also presented for the current numerical model.

The random packings of cubic or cubic particles in different cylindrical columns were generated using a glued-sphere discrete element method (DEM). The detailed packing structure and porosity distribution of the bed has been presented. Similar to spherical packings, it is found that the inter-particle voidage (\(\epsilon_p\)) decreases with the column to particle diameter ratio (\(D/d_p\)). However, \(\epsilon_p\) versus \(D/d_p\) curves are not smooth for particles with a high-aspect ratio.

The flow simulations have been performed for a wide range of macroscopic Reynolds number (\(Re_{dp} = [0.1 \ 400]\)). It is found that the microscopic Reynolds number (\(Re_{\epsilon_p}\)) for the intra-particle flows only extends to \(Re_{\epsilon_p} \approx 14\) for the highest \(Re_{dp}\) simulations. The non-dimensional pressure drop (\(f\)) versus macroscopic Reynolds number (\(Re_{dp}\)) plots follow \(f = \alpha + \beta Re_{dp}^{0.68}\). Compared to beds with solid particles where \(f \sim Re_{dp}\), particles with internal pores scaling like \(f \sim Re_{dp}^{0.68}\) have an additional advantage in pressure drop reduction for increasing flow Reynolds numbers. The flow distribution, i.e. the flow rate inside and outside the particles has been presented for different packing structures. It is found that the fluid has more tendency to flow inside the particles with increasing \(Re_{dp}\). The effect of \(\epsilon_p\) on the overall flow behavior has been also studied.

**Appendix A. Grid interdependence test**

In the current modeling approach, as we are fully resolving the interstitial space between the porous particles, it is important to check the accuracy with respect to the spatial discretization of the present results. To check the grid converge, we have taken a small section of the C30–P3 column, and performed simulations with varying grid sizes at \(Re = 500\). Three different grids, G10 (10 grid cells across the shortest length of the particle), G20 and G30 are considered; and the non-dimensional friction factor (\(f\)) for different grid resolutions are shown in Table 4. It is found the G20 grids provide grid independent results, and as a result similar grid resolutions are used for all other cases.

### Table 4

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### Table 5

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**References**


