

INCLUDING PORE CHANNEL TORTUOSITY IN ERGUN'S EQUATION USING SPHERES PACKING OF DIFFERENT SIZES

Ricardo P. Dias

Departamento de Tecnologia Química e Biológica, Escola Superior de Tecnologia e de Gestão, Instituto Politécnico de Bragança, Campus de Santa Apolónia, 5301-857 Bragança, Portugal

Email: ricardod@ipb.pt

Abstract

The experimental porosity (ϵ) measurements ranged from 0.1955 (minimum porosity region of binary packing) to 0.3974 (monodisperse packing) and compared well with different models from the literature. Using experiments at low Reynolds numbers (0.15 to 0.3: Darcy regime) the tortuosity coefficient, τ , was modelled using the formula $\tau = \left(\frac{1}{\epsilon}\right)^n$ and it was found that n is well described by $n = 0.1836 \ln(\epsilon) + 0.682$. Increasing Ergun Reynolds number (Re_{Erg}) to the range $2.87 < Re_{Erg} < 110.42$ we studied the viscous-inertial regime (non-Darcy flows) and compared the friction factor f_{Erg} experimental measurements with the model from Comiti and Renaud (1989), this model including the tortuosity coefficient. It was found that the experimental data was well described by this model using a Nikuradse friction factor $f/2 \cong 0.06$ (a roughness about 0.7 the value of the pore diameter D_H). The friction factor decreased from 0.0968 to 0.06 given that we obtained loose packings (porosity of mono-sized packings was in the range 0.3723 to 0.3974) and Comiti and Renaud obtained dense packings (mono-sized packing of spheres with $\epsilon = 0.36$). f_{Erg} experimental measurements were also compared with the model from Molerus (1980), this model including the influence of the Reynolds number in the inertial term and the packing parameter ζ . Using the experimental data from the present study (ϵ and f_{Erg}) we related ζ with the porosity, ζ being given by $\zeta = 0.9992(1 + \epsilon)^{-0.1940}$. This means that, in the present study, ζ was located in the approximate range 0.938 to 0.962, the higher values of ζ being obtained with the lower porosities (porosities around 0.2). It is reasonable to expect $\zeta \rightarrow 1$ when $\epsilon \rightarrow 0$ (the characteristic dimension of the pore space, σ , available for fluid flow tends to zero). Using the parameter ζ from the present study, the model from Molerus was compared with the model from Montillet et al. (2007) for large particle Reynolds numbers Re_p ($Re_p \leq 3000$), the model from Molerus being located between the predictions resulting from the Montillet model for dense and loose fixed beds.

Keywords: Filtration, Porosity, Darcy equation, Porous Media, Tortuosity, Ergun equation

1. Introduction

Fluid flow through the tortuous pore channels of consolidated and unconsolidated porous media is well described by the Hagen-Poiseuille's formula [1,2]:

$$\Delta P = \frac{2K_0 \mu L_{av} v}{D_H^2} \quad (1)$$

where L_{av} is the average travel distance of a fluid element in the channel with a shape factor K_0 and hydrodynamic diameter D_H . ΔP being the pressure drop of the fluid with viscosity μ and average interstitial velocity v : this velocity can be related with the Darcy velocity, u_D , through [1]:

$$v = \frac{u_D}{\varepsilon} \tau \quad (2)$$

u_D being calculated dividing the flow rate, Q , by the total cross-sectional area, A_T , of the flow; ε being the bed porosity and τ the tortuosity coefficient – the ratio $(\frac{L_{av}}{L})$ between the averaged travel distance, L_{av} , of a fluid element and the length of the bed L .

The hydrodynamic/hydraulic mean diameter of pore space can be calculated using [2, 3]:

$$D_H = 4 \frac{\text{Volume of voids filled with fluid}}{\text{Wetted surface area of the bed } (A_S)} \quad (3)$$

The volume of voids it's given by εV_T , V_T being the total volume of the pore space. The volume of particles (V_P) in the bed may be calculated by $(1-\varepsilon) V_T$. Therefore, $V_T = V_P/(1-\varepsilon)$. Hence:

$$D_H = 4 \frac{\varepsilon}{\frac{A_S}{V_T}} = 4 \frac{\varepsilon}{\left(\frac{A_S}{V_P}\right)(1-\varepsilon)} \quad (4)$$

In a packing of spheres $A_S/V_P = (\pi d_p^2/(\pi d_p^3/6)) = 6/d_p$, d_p being the spheres diameter. Hence:

$$D_H = \frac{2}{3} d_p \frac{\varepsilon}{1-\varepsilon} \quad (5)$$

In spheres packing beds of different sizes (containing spheres with diameter D and d) the average particle average, d_{av} , is calculated considering $6/d_{av} = (6/D)X_D + (6/d)(1-X_D)$, X_D being the volume fraction of spheres with large diameter D and $1-X_D$ the volume fraction of spheres with small diameter d . Introducing D_H (given by Eq. (5)) and v (given by Eq. (2)) in Eq. (1) we get:

$$\Delta P = \frac{36K_0 \tau^2 (1-\varepsilon)^2 \mu u_D L}{\varepsilon^3 d_p^2} \quad (6)$$

This formula being the Kozeny-Carman equation and $K_0 \tau^2$ the Kozeny coefficient [3]. The permeability k of a granular bed is characterized by the relation [4]:

$$k = \frac{\varepsilon^3 d_p^2}{36K_0 \tau^2 (1-\varepsilon)^2} \quad (7)$$

Darcy Law is obtained when inserting k in Eq. (6). Darcy Law holds when [5]:

$$Re_{Erg} = \frac{\rho d_p u_D}{\mu(1-\varepsilon)} < 5 \quad (8)$$

Re_{Erg} representing Ergun's Reynold number. Carman observed a decrease of permeability (transition from Darcian regime to viscous-inertial regime) as Re_{Erg} exceeded 12. Dzmitry and Tallarek [6] used the interstitial Reynolds number ($Re_{int} = \frac{\rho d_p u_D}{\mu \varepsilon}$) to demarcate different flow regimes and stated a gradual transition between Darcian regime and viscous-inertial regime when $Re_{int} > 1$. These authors emphasize that it is impossible to establish a sharp demarcation between the different flow regimes. Wang et al. [7] verified that the critical Reynolds value does not exist and quantified the transition between flow regimes using Reynolds number ranges.

Carman [3] assumed $K_0 = 2.5$ (narrow rectangular channels) and $\tau = \sqrt{2}$ for mono-sized granular beds containing spheres. Therefore, Carman found a Kozeny coefficient close to 5. Other authors [2,8] consider $K_0 = 2$ (circular cross-section and other shapes with similar K_0) and that τ varies with the porosity of the granular beds [4,9]. Mauret and Renaud (1996) [9] and Comiti and Renaud (1989) [10] considered $K_0 = 2$ and used the relation $\tau = 1 + p \ln\left(\frac{1}{\varepsilon}\right)$, p being located in the range 0.41 to 1.66. Mota et al. [4] and Dias et al. [8] used the formula $\tau = \left(\frac{1}{\varepsilon}\right)^n$ and n lied in the range 0.4 – 0.5. A value of $n = 0.5$ was found by these authors when studding mono-sized packing beds, n assuming a value of 0.4 in the regium of minimum porosity of fixed-beds containing different sized spheres. The referred authors [8] explained n variation by the distortion effect of small particles (d) arrangement occurring near de large particles (D) surface and a jamming effect [8]. Hence, the assumption that $K_0 \tau^2$ (Kozeny coefficient) is a constant is acceptable in the packing porosity range 0.36-0.4 (mono-sized packing of spheres). Beyond this range a dependence of the tortuosity on porosity must be taken in consideration and the Kozeny coefficient must be considered as a variable - Mauret and Renaud (1996) [9] presented different relations between the Kozeny coefficient and porosity.

The dependence of binary packing porosity (ε), containing binary mixtures of glass spheres (d) and (D), on particle size ratio ($\delta = d/D$) and fractional content (X_D) was studied by different authors [4,11]. A linear mixture model, which is applied for binary packing when $\delta \rightarrow 0$ is used in many works. In these works, the overall porosity is presented in the form of the product of fractional porosities: $\varepsilon = \varepsilon_D(X_D) \times \varepsilon_d(X_D)$. Using this model ($\delta \rightarrow 0$), a minimum porosity of $\varepsilon_{min} = \varepsilon_D^0 \varepsilon_d^0$ is observed at large - sized particle volume fractions $X_{D,min} = \frac{1-\varepsilon_D^0}{1-\varepsilon_D^0 \varepsilon_d^0}$. ε_D^0 and ε_d^0 correspond to the porosities of mono-sized packings containing spheres D and d , respectively. Dias et al. (2004) [11] modelled the porosity in the range $X_D \in [0, X_{D,min}]$:

$$\varepsilon = \frac{\varepsilon_d^0 (1 - X_D) \exp\left(1.2264 X_D^{\frac{1}{\sqrt{\delta}}}\right)}{1 - \varepsilon_d^0 X_D} \quad (9)$$

For mixtures enriched with small particles (d), where $X_D < 0.65$, the amount of large particles (D) is not enough to build up a support skeleton inside the mixture. Therefore, the mixing procedure does not affect the individual porosities ε_D^0 and ε_d^0 . The region of minimum porosity ($0.65 < X_D < 0.75$) correspond to “flooded beds” – small particles are able to wedge between large particles of the skeleton and the overall bed porosity may increase. However, when the ratio δ is small enough to allow small particles to penetrate freely into the skeleton void, the skeleton is kept fixed till $X_{D,min}$: in this δ region the exponential term from Eq. (9) tends to 1 and the overall porosity is described by the ideal filling mechanism (wall effect between the different sized particles and wedging effect are negligible) [11].

As referred previously, Carman [3] observed a decrease of permeability (transition to non-Darcy flows) as Re_{Erg} exceeded 12. Using a Forchheimer type equation Carman modelled the pressure drop in the Re_{Erg} region $0.06 < Re_{Erg} < 60\,000$:

$$f_{Erg} = \frac{\Delta P d_p \varepsilon^3}{L \rho u_D^2 (1 - \varepsilon)} = \frac{A}{Re_{Erg}} + \frac{B}{Re_{Erg}^C} \quad (10)$$

f_{Erg} being the Ergun friction factor [12]. Using beds of spherical particles, Carman found $A = 180$, $B = 2.871$ and $C = 0.1$ (porosity was in the range 0.3 to 0.42). One can see that when the flow velocity is very small Eq. (10) simply reduces to Darcy Law (Eq. (6)), while as the fluid velocity increases (viscous- inertial regime), Eq. (10) predicts a deviation from the linear relationship between pressure drop and flow velocity, and at very high velocities (turbulent regime – chaotic inertial flow) the second term on the right-hand side of Eq. (10) is dominating [6]. A value of $A = 180$ in Eq. (10) means that, in Eq. (6), $36K_0 \tau^2 = 180$ and, therefore, a Kozeny coefficient equal to 5. Brauer (1971) [13], also using spherical particles, found a similar equation in the Re_{Erg} region $0.01 < Re_{Erg} < 40\,000$ and proposed $A = 160$, $B = 3.1$ and $C = 0.1$. Hence, the Kozeny coefficient was equal to 4.44 in the referred study [13].

Using spheres, coke/coal, sand, cylinders and tablets, Ergun (1952) [12] found a friction factor also described by Eq. (10). The measurements were in the range $1 < Re_{Erg} < 2400$. In this study $A = 150$, $B = 1.75$ and $C = 0$. Allen et al. (2013) [14] state that this equation over-predicts the pressure drop for randomly packed beds of smooth spheres and that it may not be applicable when $Re_{Erg} > 500$. These authors suggested that correlations with a constant C different from zero in Eq. (10) predict better pressure drop data in the inertial regime. Hicks (1970) [15] also stated that the Ergun equation does not fit the pressure drop data for smooth spheres for values of $Re_{Erg} > 500$ and found $C = 0.2$. However, the different correlations (with $C = 0$ or $C \neq 0$), as Re_{Erg} decreases bellow 500, tend to converge [14]. After testing the Ergun equation using many more data than ever before [6], Macdonald et al. (1979) [16] found that the following values of A and B give the best fits to all of the involved data: $C = 0$, $A = 180$ and $B = 1.8$ (smooth particles) or $B = 4.0$ (rough particles). These authors also

concluded that using $\varepsilon^{3.6}$, instead of ε^3 in the Ergun equation, would result in a better fit to data points and related A with ε . Using parallelepipedal particles, such as wood chips used in paper pulp making industry, Comiti and Renaud (1989) [10] found $C = 0$, $A = 72\tau^2$ and $B = 0.5808\tau^3$. In order to find A and B , these authors [9, 10] used the tortuosity definition $\tau = 1 + p \ln\left(\frac{1}{\varepsilon}\right)$. The explicit introduction of tortuosity in Eq. (10) highlights the fact that the fluid particles travel a distance $L_{av} - L$ in the transverse direction.

2. Materials and methods

The soda-lime glass spheres used on the pressure drop measurements were obtained from Sigmund Lindner and they had the following average diameters: 1.2×10^{-4} m, 2.5×10^{-4} m, 3.375×10^{-4} m, 8.75×10^{-4} m, 1.125×10^{-3} m and 5×10^{-3} m. The density of the glass beads was 2500 kg/m^3 .

The glass spheres with two different diameters (d and D) were mixed using a water-glycerol solution as the binder between the different sized particles [17]. The mixture was transferred to a square acrylic transparent column (please see Fig. 1) and the column was tapped vertically until we got no variation on the packing thickness. The column used in the experiments had 0.08 m on the inner side and 0.4 m length. A square wire cloth from Haver & Boecker with aperture with 3.2×10^{-5} m was used as a layer support. After that, bed porosity (ε) was determined using gravimetry - packing height (L) varied between 10 and 15 cm. The pressure loss (ΔP) of the constructed binary packing beds was determined measuring the flow rate (Q) at a fixed height of water – the experimental setup being described in a previous investigation [18]. In order to obtain different Reynolds numbers, two heights of water were used: 109 cm and 182 cm. The cross-sectional area of the prismatic column and Q allowed the calculation of the Darcy velocity (u_D).

Experiments at low Re_{Erg} numbers (0.15 to 0.30) allowed permeability calculation. The permeability, k , of the constructed binary packing beds (mixing spheres with diameter $d = 1.2 \times 10^{-4}$ m and spheres with diameter $D = 5 \times 10^{-3}$ m at different volume fractions X_D) was determined by Eq. (11) [5]:

$$k = \frac{L}{\left(\frac{\Delta P}{\mu u_D} - R_M\right)} \quad (11)$$

R_M was determined in a experiment without bed ($L = 0$), being equal to $2.3250 \times 10^8 \text{ m}^{-1}$.

In these experiments, the volume fraction, X_D , of spheres with diameter $D = 5 \times 10^{-3}$ m was located in the range $0 \leq X_D \leq 0.65$ and the porosity was in the range $0.1955 \leq \varepsilon \leq 0.3924$, 0.3924 being the porosity of the mono-sized packing ($X_D = 0$) and 0.1955 being the porosity of the binary packing with $X_D = 0.65$.

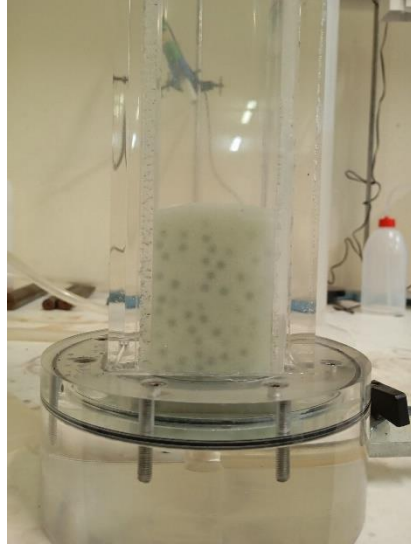


Figure 1: Binary packing containing 25 % of 5 mm (diameter D) glass spheres and 75 % 0.25 mm (diameter d) glass spheres ($\varepsilon = 0,342$).

These experiments allowed the calculation of the tortuosity coefficient, τ . Using the experimental values of k , ε , $K_0 = 2$ and Eq. (7) it was possible to determine the tortuosity coefficient. The values of τ , ε and the tortuosity definition $\tau = \left(\frac{1}{\varepsilon}\right)^n$ allowed the calculation of n . As in previous investigations [8,18,19] n was located in the approximate range 0.4 (region of minimum porosity, $X_D = 0.65$) to 0.5 (mono-sized packing bed, $X_D = 0$). In Eq. (7) the average particle diameter (d_p or d_{av}) was calculated using $1/d_{av} = X_D / D + (1-X_D) / d$ [19].

In the remaining experiments the Reynolds number Re_{Erg} varied between 2.87 and 110.42 since the diameter of the small spheres d was increased ($2.5 \times 10^{-4} \text{ m} \leq d \leq 1.125 \times 10^{-3} \text{ m}$). The diameter of the large particles ($D = 5 \times 10^{-3} \text{ m}$) was maintained in these latter experiments. In this Re_{Erg} range, we observed the transition to non-Darcy flows: deviation from the linear relationship between pressure drop and flow velocity. Therefore, in this Re_{Erg} range, Eq. (10) was used to model the experimental data. Coefficient A from Eq. (10) requires the knowledge of the tortuosity coefficient, τ . The tortuosity coefficient can be estimated using $\tau = \left(\frac{1}{\varepsilon}\right)^n$, n being described by [19]:

$$n = 0.5 - \lambda F_D \quad (12)$$

F_D is the fraction of surface area of large particles D . The decreasing of n from 0.5 to 0.4 when X_D approach the region of minimum porosity may therefore be explained by the increase of the total surface area of large particles present in the mixtures and thus by the increase of the fraction of porous media involved in the “wall effect” between the different sized particles [19]). F_D was described by [19]:

$$F_D = \frac{X_D}{X_D + (1-X_D)\left(\frac{D}{d}\right)^2} \quad (13)$$

λ was described by [19]:

$$\lambda = 0.062(D/d - 1) \quad (14)$$

The authors [19] found that the experimental values of n were well described by Eq. (12). Since Eq. (12) depends on X_D and $D/d = \delta^{-1}$, in this work we develop an equivalent relationship between n and ε (which simplifies the use of the definition $\tau = \left(\frac{1}{\varepsilon}\right)^n$).

3. Results and discussion

Experiments at low Re_{Erg} numbers (0.15 to 0.30) allowed n calculation. These experiments were carried out mixing spheres with diameter $d = 1.2 \times 10^{-4}$ m with spheres having diameter $D = 5 \times 10^{-3}$ m ($D/d = \delta^{-1} = 41.67$). The volume fraction of large spheres with diameter $D = 5 \times 10^{-3}$ m was located in the range $0 \leq X_D \leq 0.65$. Using Eq. (7) and the tortuosity definition $\tau = \left(\frac{1}{\varepsilon}\right)^n$, the values of n were calculated from known porosity, average particle size and permeability [8,19]:

$$n = \frac{\ln\{36kK_0(1-\varepsilon)^2/(\varepsilon^3 d_{av}^2)\}}{2\ln(\varepsilon)} \quad (15)$$

As it was found in previous investigations [8, 19], the new experimental data shows that n is not a constant (please see Fig. (2)) and lies in the approximate range 0.4 and 0.5 for the investigated loose packings. The reduction of n when the system approaches minimum porosity ($n = 0.3924$ for the lower porosity $\varepsilon = 0.1955$) can be explained with the small size particle packing distortion in the regions close to large particles surface and particles jamming in the void of large particles skeleton. These distortion effects vanish as X_D approaches zero, n assuming a value close to 0.5 for $X_D = 0$ (ε of mono-sized packing was 0.39237).

In previous investigations [8,19], n and ε were both related with X_D and D/d , Eq. (12) and Eq. (9), respectively. Therefore, using only the porosity experimental data it is possible to describe the behaviour of n (please see Fig. (2) for $D/d = 41.67$). n is well described by ε ($R^2 = 0.9473$) using the following logarithmic expression:

$$n = 0.1795 \ln(\varepsilon) + 0.6829 \quad (16)$$

The present n experimental together with the n experimental data from previous investigations [8, 19] (please see Fig. (3)) is well described by a similar relation ($R^2 = 0.9370$):

$$n = 0.1836 \ln(\varepsilon) + 0.6821 \quad (17)$$

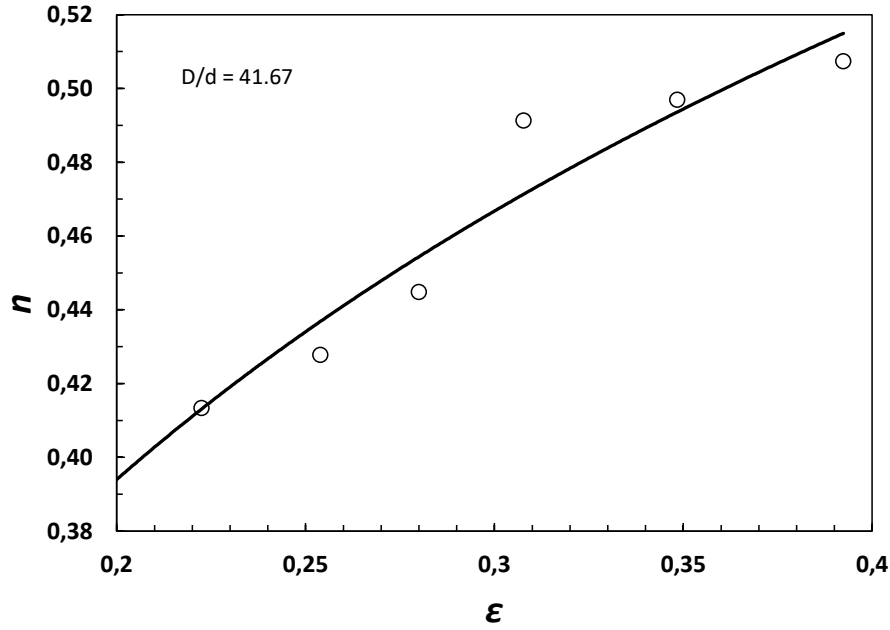


Figure 2: Binary packings containing 5 mm (diameter D) glass spheres and 0.12 mm (diameter d) glass spheres. Using volume fractions X_D in the range $0 \leq X_D \leq 0.65$, n was estimated using Eq. (15).

We prefer the latter relationship since it was obtained with more experimental data points and R^2 is similar. This relationship between n and ϵ (Eq. (17)) has a wider and easier application compared to Eq. (12), since it does not include the variables X_D and D/d in its definition.

Using the same experimental data to analyse a different, and also widely used, tortuosity definition $\tau = 1 + p \ln\left(\frac{1}{\epsilon}\right)$ [2, 9, 10] we found that p varies in the approximate range 0.527 (region of minimum porosity of binary packings, $X_D = 0.65$) and 0.647 (mono-sized packings, $X_D = 0$, with porosities in the range 0.37 to 0.397: loose packings). The p -values obtained in the present study fall within the range reported by Comiti and Renaud (1989) [10].

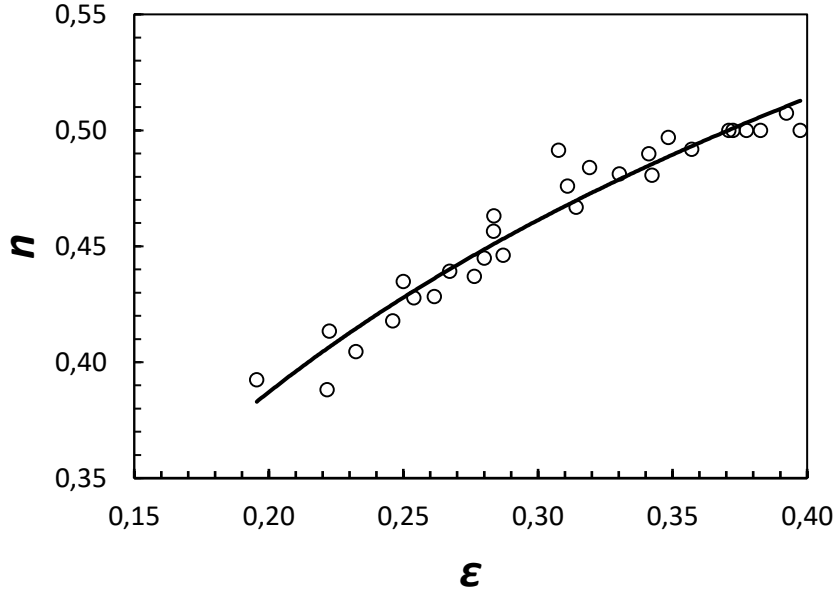


Figure 3: Using n experimental data from previous investigations [8, 19] and n experimental data from present investigation (please see Fig. (2)) we found $n = 0.1836 \ln(\varepsilon) + 0.6821$.

Analysing the experimental Kozeny coefficients ($K_0 \tau^2$) reported by Wyllie and Gregory (1955) [20] and using a shape factor K_0 equal to two, it is possible to estimate the tortuosity related to these experiments. Since the aforementioned authors provide the porosity of the used monodisperse and binary packings, it is possible to estimate the values of n ($n = \ln(\tau) / \ln(\frac{1}{\varepsilon})$) and p ($p = (\tau - 1) / \ln(\frac{1}{\varepsilon})$), with the resulting values of n varying in the range 0.379 to 0.483 and the values of p in the range 0.530 to 0.607. Values of n close to 0.5 correspond to monodisperse loose packings (ε was in the range 0.368 to 0.4015) and values of n close to 0.4 (0.379) correspond to a binary packing with a porosity of 0.1868 [20]. It is interesting to note that the referred binary mixture was constructed in the region of minimum porosity ($X_D = 0.7$), where the distortion effects mentioned earlier (wall effect and jamming) are at their maximum. Thus, the n results obtained in the present study agree well with the experimental values of Wyllie and Gregory [20]. Using Wyllie and Gregory [20] experimental data, p -values of 0.607 were obtained with mono-sized loose packings (ε was in the range 0.368 to 0.4015) and p -values of 0.530 were obtained with a binary mixture in the region of minimum porosity ($\varepsilon = 0.1868$; $X_D = 0.7$): this variation compares well to the present study (p ranged between 0,527 and 0.647 in the present study).

In Fig. (4) the porosity experimental data from the present study is compared to a model from the literature (Eq. (9)): maximum deviation is 4.47 % and average deviation 1.77%. ε_d^0 varied in the range 0.371 to 0.397; X_D was located in the range $0 \leq X_D \leq 0.69$ and $D/d = \delta^{-1}$ was in the range 4.44 to 41.67. The experimental data for $D/d < 10$ also compares well to the model from Mota et al. (2001) [4] - maximum deviation is 5.49 % and average deviation 2.08 %. These comparisons highlight the good quality of the measurements.

To increase the Re_{Erg} number ($2.87 < Re_{Erg} < 110.42$), and using the same water heights, the diameter of the smaller spheres was increased, d being located in the

range $2.5 \times 10^{-4} \text{ m} \leq d \leq 1.125 \times 10^{-3} \text{ m}$. The diameter of the larger spheres D was maintained and was equal to $5 \times 10^{-3} \text{ m}$. In this Re_{Erg} range, we observed the transition to non-Darcy flows – deviation from the linear relationship between pressure drop and flow velocity. Therefore, in this Re_{Erg} range, Eq. (10) was used to model the experimental data. Coefficient A from Eq. (10) requires the knowledge of the tortuosity coefficient, τ . The tortuosity coefficient can be estimated using $\tau = \left(\frac{1}{\varepsilon}\right)^n$, n being described by Eq. (17):

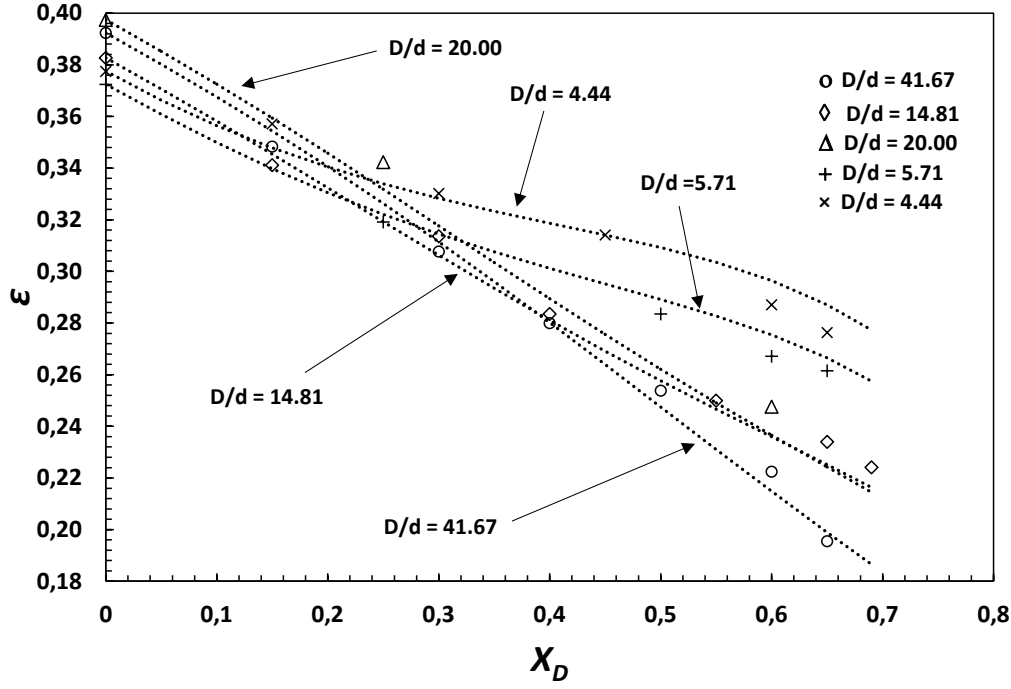


Figure 4: Porosity (ε) experimental data used in the experiments for different values of volume fraction X_D and different size ratios $D/d = \delta^{-1}$. The experimental data (dots) is compared with Eq. (9) [11]- lines.

From the viewpoint of fixed-beds percolation, tube flow is a singular case. With laminar tube flow, the fluid elements move rectilinearly and without acceleration, i.e. inertia effects are absent. This is the reason why laminar flow is an exact solution of the Navier-Stokes equations for any Reynolds number and why inertia effects are observed for tube flow only on the onset of turbulence, so that a critical Reynolds number is observed. As a consequence of the frequent and intense deflections of fluid flow inside a packing, inertia effects dominate long before the onset of true turbulence [21]. These considerations justify the observation (please see Fig. (5)) of inertial effects for low Reynolds numbers ($Re_{Erg} \geq 2.87$). In this figure we present the experimental data from the friction factor f_{Erg} for different Reynolds numbers Re_{Erg} .

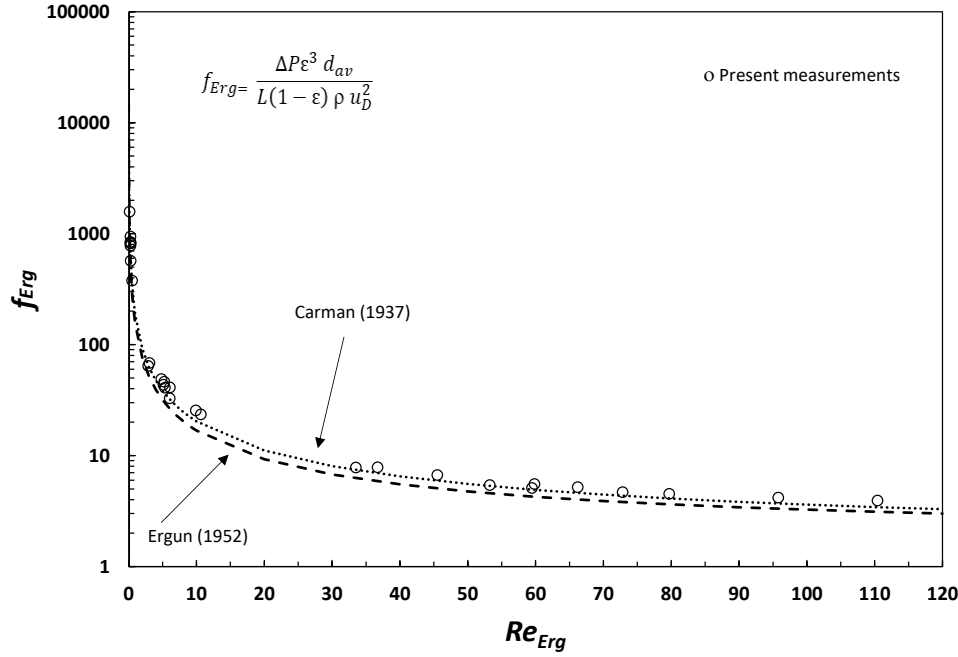


Figure 5: f_{Erg} experimental data for different Reynolds numbers Re_{Erg} . The friction factor f_{Erg} was calculated using Eq. (10) and the Reynolds number Re_{Erg} was determined using Eq. (8).

Fig. (5) shows that the experimental data are better described using the Carman (1937) model [3] compared to the Ergun (1952) model [12]: average deviation between the model from Carman and experimental measurements is 16.78%. In either case, the experimental data are superior to the aforementioned models, since these do not include tortuosity (as a variable) in their definition. Carman's model includes $C = 0.1$, while Ergun assumed $C = 0$. Carman assumed $A = 180$, whereas in the present study $A = 36K_0 \tau^2$ which results in obtaining values of A greater than 180. A value of $A = 180$ in Eq. (10) means that, in Eq. (6), $36K_0 \tau^2 = 180$ and, therefore, a Kozeny coefficient equal to 5. In the present study the Kozeny coefficient was ≥ 5.16 and, therefore, A was located in the range 185.59 (mono-sized packing with $\tau = 1.6055$, $n = 0.5125$ and $\varepsilon = 0.397$) to 250.90 (a binary packing with $\tau = 1.8668$, $n = 0.3924$ and $\varepsilon = 0.1955$). Hence, considering only mono-sized fixed beds (porosity was in the range 0.372 to 0.397), we may observe a good agreement between the model from Carman and experimental data from the present study (please see Fig. (5)). In this case (six different mono-sized fixed beds) the average deviation between the model from Carman and f_{Erg} experimental measurements was 3.52 %. The experimental data are significantly higher than the Ergun model, given that this model includes a constant $A = 150$: average deviation between the model from Ergun and experimental measurements is 37.04 %.

Taking these considerations into account, in a first approach, we studied Eq. (10) using $C = 0$. Having in mind that $A = 36K_0 \tau^2$, and admitting $K_0 = 2$ [2, 4, 8, 10, 19], we get:

$$f_{Erg} = \frac{\Delta P d_p \varepsilon^3}{L \rho u_D^2 (1-\varepsilon)} = \frac{72 \tau^2}{Re_{Erg}} + B \quad (18)$$

Considering the obtained f_{Erg} experimental data, Re_{Erg} described by Eq. (8) and the tortuosity defined by $\tau = \left(\frac{1}{\varepsilon}\right)^n$, n being described by Eq. (17), we then use a linear regression analysis where f_{Erg} represents the y-axis and $\frac{\tau^2}{Re_{Erg}}$ the x-axis, we get $y = 71.8840 \times x + 1.9431$, R^2 being equal to 0.99 (please see Fig. (6)). Therefore, in the present study, constant B from Eq. (10) assumed an average value $B = 1.94$, this value being higher than the constant suggested by Ergun (1.75) [12]. Macdonald et al. (1979) [16] found $B = 1.8$ for smooth particles and $B = 4.0$ for rough particles.

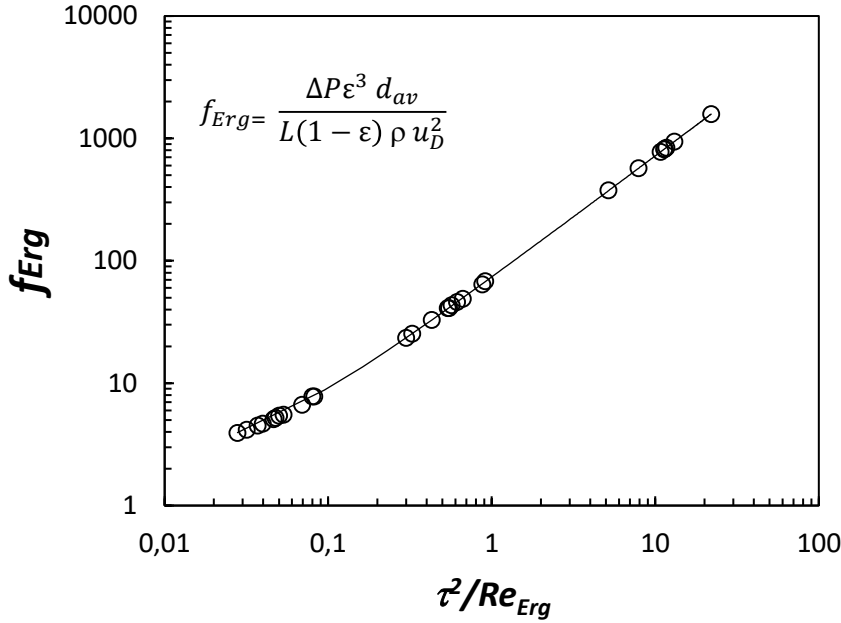


Figure 6: f_{Erg} experimental data for different τ^2 / Re_{Erg} numbers. The line represents the equation $f_{Erg} = 71.8840 \times \tau^2 / Re_{Erg} + 1.9431$ ($R^2 = 0.99$).

Given that Re_p is described by $Re_p = \frac{\rho d_p u_D}{\mu}$ we can rewrite Eq. (18):

$$f_{Erg} = \frac{\Delta P d_p \varepsilon^3}{L \rho u_D^2 (1 - \varepsilon)} = \frac{71.8840 \tau^2 (1 - \varepsilon)}{Re_p} + 1.9431 \quad (19)$$

In Fig. (7) we present the f_{Erg} described by Eq. (19) in two limiting cases: $\varepsilon = 0.1955/\tau$ and $\varepsilon = 0.397/\tau$. Given the good correlation between Eq. (19) and the experimental results, f_{Erg} experimental data fall between the two theoretical limit curves (please see Fig. (7)).

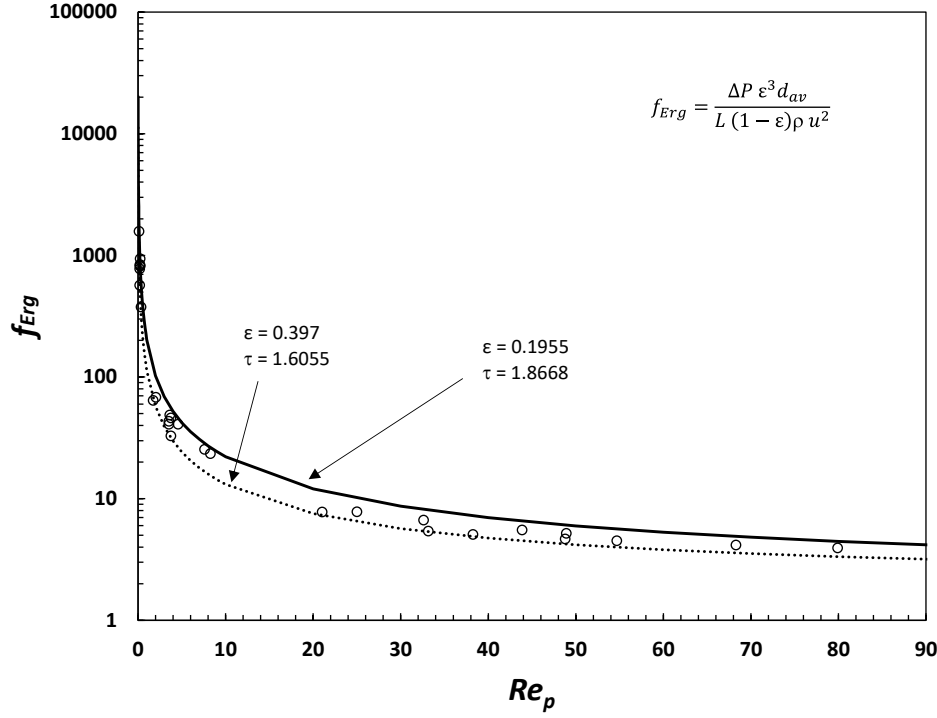


Figure 7: f_{Erg} experimental data for different Reynolds numbers Re_p . The experimental data are compared with Eq. (19) in two limiting cases: $\varepsilon = 0.1955/\tau = 1.8668$ and $\varepsilon = 0.397/\tau = 1.6055$.

Ergun constant B is dependent from packing porosity [22], packing porosity and turbulence promoters [23], tortuosity and pores roughness [10]. Tobis [23] has shown, by inserting obstacles into flow passages between spheres in a simple cubic arrangement that the constant 1.75 in the Ergun equation can vary by a factor up to almost five, depending on fixed-bed porosity and the alignment/shape of the obstacle [23, 14]. In order to take in account numerous direction changes, Comiti and Renaud [10] assumed that the tortuous pores are very rough conduits, the roughness of which, E , has the same range of magnitude as the pore diameter, D_H . Taking into account the previous considerations, the constant of 1.9431 presented in Eq. (19) is the average of different values of B generated by packings with different porosity and tortuosity. Next, using the Comiti and Renaud model [10], we analysed the influence of tortuosity and roughness on the constant B of Eq. (18). The term corresponding to kinetic-energy losses was written by these authors as $B = 6(\frac{f}{2})\tau^3$. By analogy with the case of cylindrical pipes, the friction factor ($f/2$) was calculated using the Nikuradse formula [9, 10]:

$$\frac{1}{\sqrt{f/2}} = 2.46 \ln \left(\frac{D_H}{2E} \right) + 4.92 \quad (20)$$

The mentioned authors performed experiments in the range $2 < Re_p < 150$ and found a good agreement between the model (Eq. (18) with $B = 6(\frac{f}{2})\tau^3$) and experimental data (mono-sized packing of spheres with $\varepsilon = 0.36$). In order to compare the model with experimental data, the authors considered a roughness "approximately equal to the pore diameter, D_H " ($E \cong D_H$ in Eq. (20)). Consequently, the authors used $f/2 \cong 0.0968$

[10]. These authors [9, 10] estimated the tortuosity using the definition $\tau = 1 + p \ln \left(\frac{1}{\varepsilon} \right)$ and considered $p = 0.41$ [10] and, later [9], $p = 0.49$ (mono-sized packing of spheres). Using the model from Molerus (1980) [21], Mauret and Renaud [9] validated the capillary model from Comiti and Renaud model [10] up to a $Re_p = 1000$. In the case of low Re_p numbers ($Re_p < 1$), the referred authors [9] suggested that the capillary model may be used for porosities smaller than 0.7. These authors also tested the capillary model against experimental data from literature: here again good agreement was found at high Reynolds numbers and strong scatter in the region of low Reynolds numbers [9].

In Fig. (8) we compare the f_{Erg} experimental data with the model from Comiti and Renaud [10] using different tortuosity definitions. Using $\tau = 1 + 0.49 \ln \left(\frac{1}{\varepsilon} \right)$ [9], the average deviation between experimental data and the referred model is 12.84% (experimental data lies slightly above the model – please see Fig. (8)). Using $\tau = \left(\frac{1}{\varepsilon} \right)^n$ with n being described by the formula found in the present work (Eq. (17)) the average deviation between experimental data and the referred model is 8.12 % (experimental data lies slightly below the model). In order to correct these deviations, we found that a value of $f/2 \cong 0.06$ (a roughness about 0.7 the value of the pore diameter D_H) describe better the experimental data: average deviation 1.82% (please see Fig (9)). In this later comparison we used $\tau = \left(\frac{1}{\varepsilon} \right)^n$, n being described by Eq. (17).

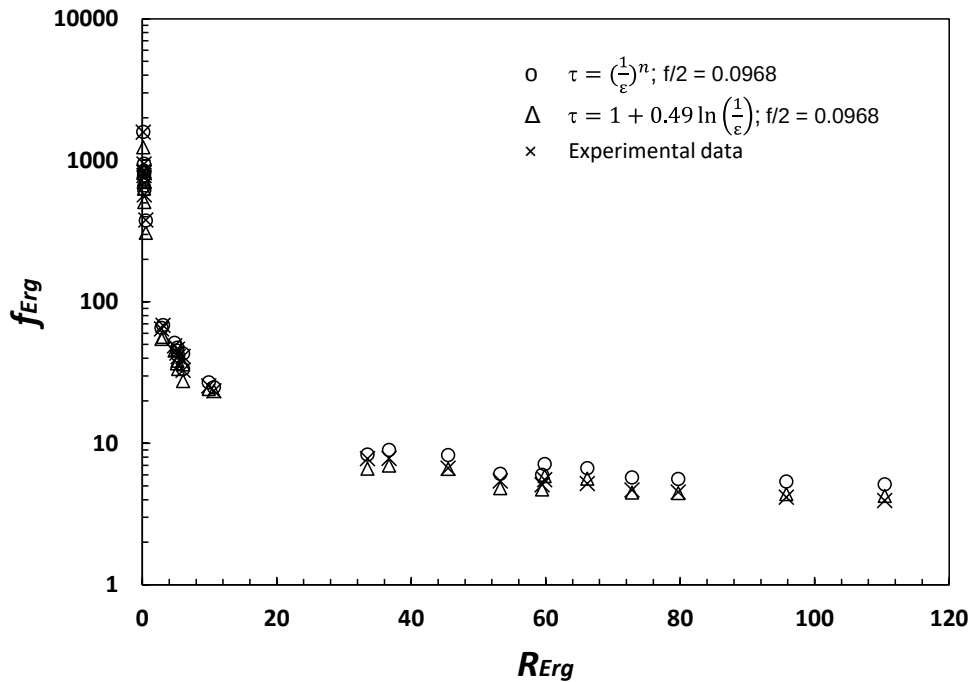


Figure 8: f_{Erg} experimental data for different Reynolds numbers Re_{Erg} . Comparison with the model from Comiti and Renaud [10] using different tortuosity definitions and $f/2 = 0.0968$ ($E \cong D_H$).

Comiti and Renaud [10] used mono-sized packings with porosity (0.36) somewhat lower than the porosity from the mono-sized packings used in present study (0.372 to 0.397). This means that Comiti and Renaud [10] obtained denser packings and this difference on packing density may justify the decrease of $f/2$ down to a value of 0.06. The Comiti and Renaud model [10] describes very well the experimental measurements from the present study, namely the influence of tortuosity (and porosity) variation on the constants A and B .

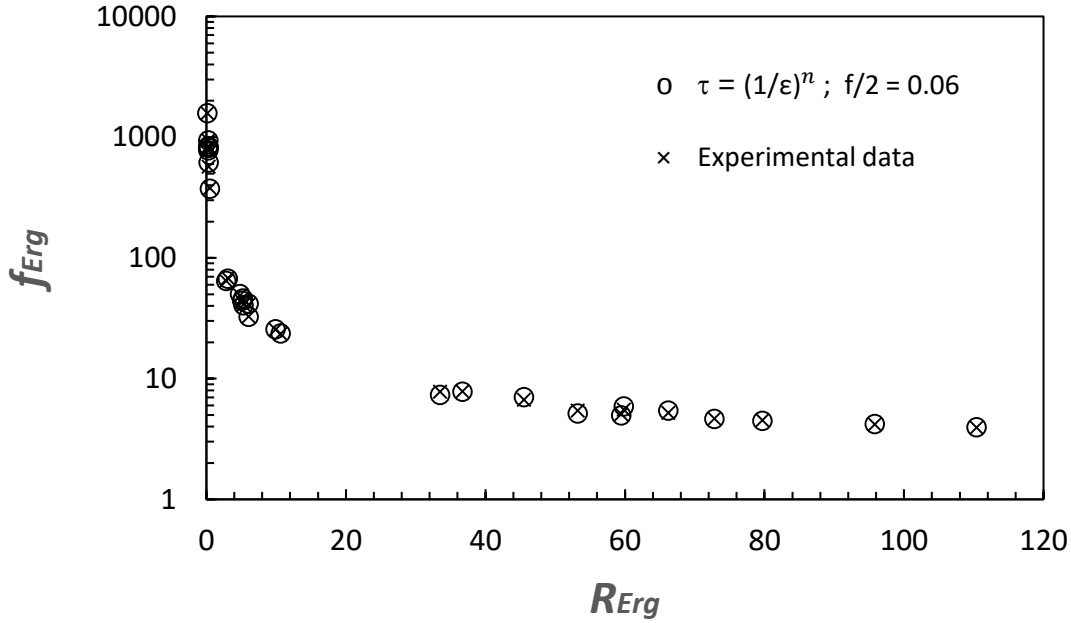


Figure 9: f_{Erg} experimental data for different Reynolds numbers Re_{Erg} . Comparison with the model from Comiti and Renaud [10] using $\tau = \left(\frac{1}{\epsilon}\right)^n$ and $f/2 = 0.06$ ($E \cong 0.7D_H$).

Using the drag coefficient of a single sphere, Molerus [21] estimated the average force (Euler number) exerted by the fluid in one particle. Writing the relationship found by Molerus in terms of f_{Erg} :

$$f_{Erg} = \frac{18\epsilon^2}{Re_p} \left\{ 1 + 0.777 \left[\frac{r_0}{\sigma} + \frac{1}{2} \left(\frac{r_0}{\sigma} \right)^2 \right] \right\} + \frac{3\epsilon^{1.5}}{Re_p^{0.5}} + 0.3\epsilon + 0.38325 \frac{r_0}{\sigma} \epsilon \quad (21)$$

This relation was confirmed by experiments up to $Re \leq 200$ ($0.36 \leq \epsilon \leq 0.64$), the Reynolds number being defined by $(\rho d_p u_D) / (\epsilon \mu)$. Molerus defined a cubic cell model with volume $V_{cell} = \zeta_1 (r_0 + \sigma)^3$ containing a solid particle with volume $V_p = \zeta_2 r_0^3$ ($\zeta_2 = (4\pi)/3$ for spheres). Considering that porosity is given by $\epsilon = (V_{cell} - V_p) / V_{cell}$ and $\zeta = \sqrt[3]{\zeta_2 / \zeta_1}$, $\frac{r_0}{\sigma}$ can be calculated using the expression:

$$\frac{r_0}{\sigma} = \frac{1}{\frac{\zeta}{(1-\epsilon)^{1/3}} - 1} \quad (22)$$

r_0 being the particle radius and σ the characteristic dimension of the pore space available for fluid flow. Using experiments at low Reynolds interstitial numbers ($Re_{int} \leq 1$) Molerus [21] found $\zeta = 0.95$ (for $\varepsilon = 0.5$). In Fig. (10) we compare our f_{Erg} experimental data with the model from Molerus using $\zeta = 0.94$ and $\zeta = 0.96$. The average deviation between the experimental data and the referred model is 28.50 % and 28.21%, respectively, and it is possible to see in Fig. (10) that the experimental data follows the trend from Eq. (21) and is located between the predictions for $\zeta = 0.94$ and $\zeta = 0.96$. In Fig. (11) we compare the f_{Erg} experimental data with Eq. (21) using $\zeta = 0.95$ (the value suggested by Molerus), the average deviation decreasing to 18.72%. Molerus suggested that ζ can be determined with sufficient accuracy from fluidized-bed expansion measurements as well as from fixed-bed pressure drop measurements and that ζ should be located in the range $\zeta \rightarrow 1$ [21]. Therefore, we used the f_{Erg} experimental data to do so. In Fig. (12) we plot the behaviour of ζ against ε . It is reasonable to expect $\zeta \rightarrow 1$ when $\varepsilon \rightarrow 0$ ($\sigma \rightarrow 0$). Hence, we added that boundary condition to the experimental data (please see Fig. (12)) and related ζ with ε (please see Eq. (23): the correlation coefficient R^2 was 0.99. Including Eq. (23) in the model from Molerus (Eq. (21)) the average deviation between the model and experimental measurements is 2.77%. Therefore, the accuracy from the Molerus model [21] is at the same level of the Comiti and Renaud model [9, 10] but has the advantage of including the influence of the Reynolds number on the inertial term ($C \neq 0$ in Eq. (10)). Different authors [3,14,13,22] suggest that correlations with $C \neq 0$ predict better pressure drop data in the inertial regime.

$$\zeta = 0.9992(1 + \varepsilon)^{-0.1940} \quad (23)$$

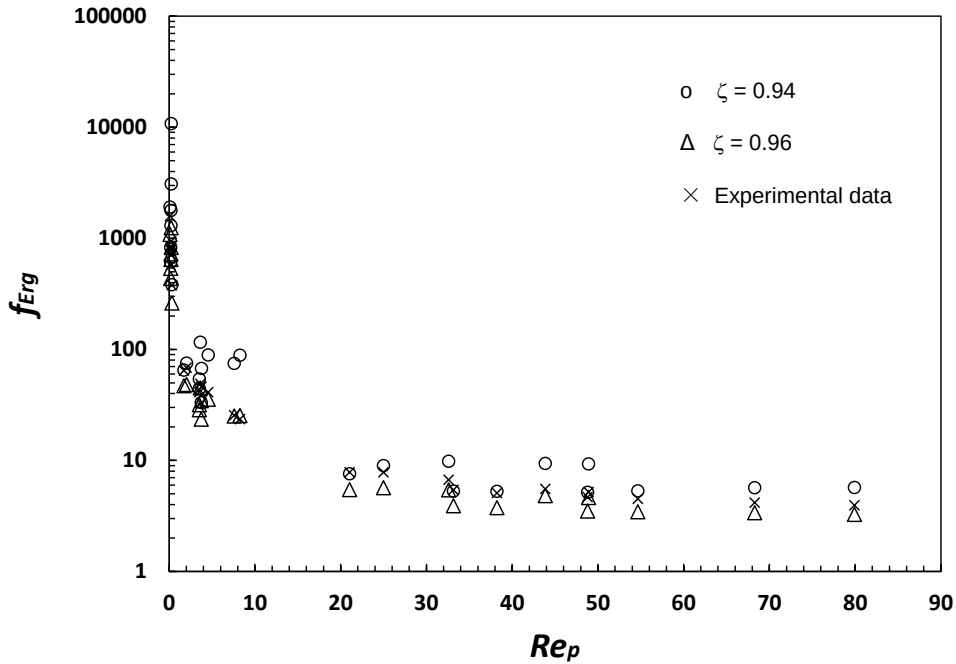


Figure 10: f_{Erg} experimental data for different Reynolds numbers Re_p . Comparison with the model from Molerus using $\zeta = 0.94$ and $\zeta = 0.96$.

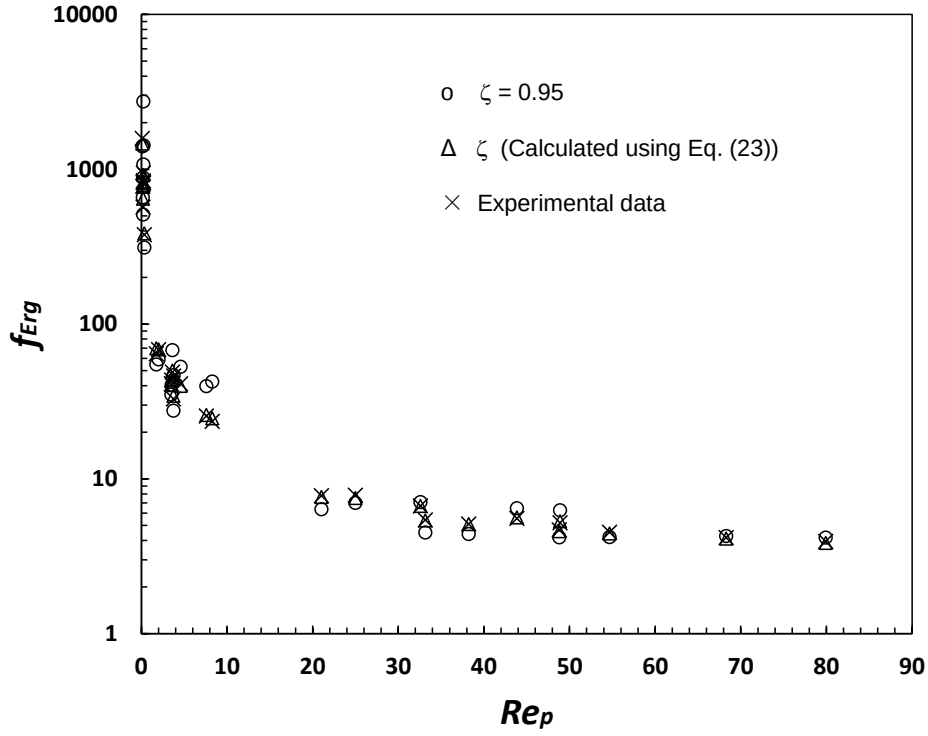


Figure 11: f_{Erg} experimental data for different Reynolds numbers Re_p . Comparison with the model from Molerus using $\zeta = 0.95$ and ζ described by Eq. (23).

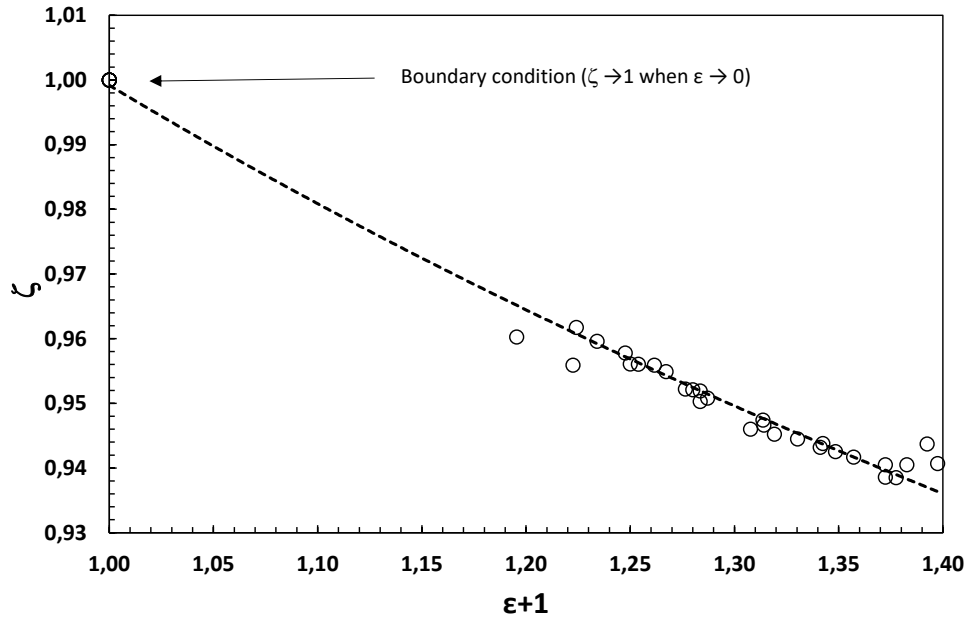


Figure 12: Relation between ζ and porosity (ϵ): $\zeta = 0.9992(1 + \epsilon)^{-0.1940}$. Using f_{Erg} experimental data, ζ was calculated as suggested by Molerus [21].

In Fig. (13) we compare the model from Molerus [21] with the model from Montillet et al. (2007) [24] (please see Eq. (24)), at large Reynolds numbers ($Re_p \leq 3000$), using a porosity typical from mono-sized fixed beds $\epsilon = 0.38$. Both studies include a

dependence of the type $Re_p^{-0.5}$ in the inertial regime. The parameter $\frac{r_0}{\sigma}$ (please see Eq. (22)) was estimated using the equation found in the present work (Eq. (23)) and it is equal to 0.9387. In Fig. (13) it can be observed that the Molerus model is located between the predictions resulting from the Montillet model for dense and loose fixed beds (dotted lines), confirming that the value of $\frac{r_0}{\sigma}$ was well estimated through Eq. (23) and the validity of the Molerus model for high Reynolds values. For large bed geometry aspect ratios, the model from Montillet et al. [24] it is given by:

$$f_{Erg} = 2.2a[1000Re_p^{-1} + 60Re_p^{-0.5} + 12] \quad (24)$$

The parameter a being equal to 0.061 and 0.05 for dense and loose packings, respectively. The authors [24] compared Eq. (24) to the Ergun equation [12] and also found that this latter equation overpredicts the pressure drop in the inertial regime. A similar conclusion may be found in the studies from Allen et al. [14] and Hicks [15].

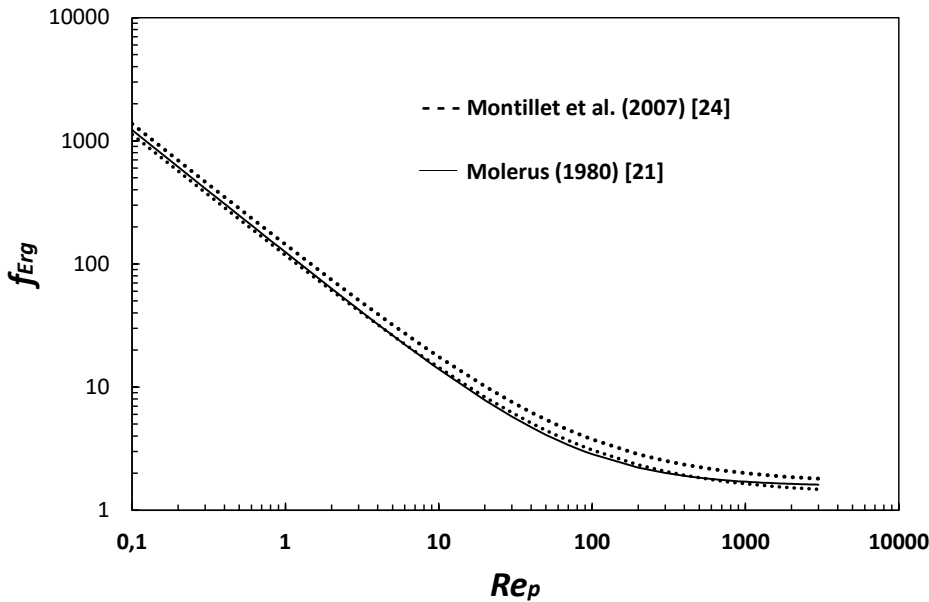


Figure 13: Relation between f_{Erg} and Re_p using the model from Montillet et al. [24] and the model from Molerus [21] for $\varepsilon = 0.38$ (mono-sized fixed bed).

4. Concluding remarks

Using experiments at low Re_{Erg} numbers (0.15 to 0.3: Darcy regime) the tortuosity was modelled using the formula $\tau = \left(\frac{1}{\varepsilon}\right)^n$ and it was found that n is well described by $n = 0.1836 \ln(\varepsilon) + 0.682$. Increasing the Reynolds number to the range $2.87 < Re_{Erg} < 110.42$ we studied the viscous-inertial regime (non-Darcy flows) and compared the f_{Erg} experimental measurements with the model from Comiti and Renaud [10]. We found that the experimental data was well described by this model using a Nikuradse friction factor $f/2 \cong 0.06$ (a roughness about 0.7 the value of the pore diameter D_H). The friction factor decreased from 0.0968 to 0.06 given that we obtained loose packings (porosity,

ε , of mono-sized packings was in the range 0.372 to 0.397) and Comiti and Renaud obtained dense packings (mono-sized packing of spheres with $\varepsilon = 0.36$). f_{Erg} experimental measurements were also compared with the model from Molerus [21], this model including the influence of the Reynolds number in the inertial term. Using the experimental data from the present study (ε and f_{Erg}) we related the parameter ζ with the porosity, ζ being given by $\zeta = 0.9992(1 + \varepsilon)^{-0.1940}$. This means that, in the present study, ζ was located in the approximate range 0.938 to 0.962, the higher values of ζ being obtained with the lower porosities (porosities around 0.2). It is reasonable to expect $\zeta \rightarrow 1$ when $\varepsilon \rightarrow 0$ (the characteristic dimension of the pore space, σ , available for fluid flow tends to zero). Using the parameter ζ from the present study, the model from Molerus [21] was compared with the model from Montillet et al. (2007) [24] for large Re_p numbers ($Re_p \leq 3000$), being located between the predictions resulting from the Montillet model for dense and loose fixed beds.

Appendix A. Nomenclature

a	parameter of Eq. (24)
A	parameter of Eq. (10)
A_T	Cross sectional-area (m ²)
A_s	wetted surface area of the bed (m ²)
B	parameter of Eq. (10)
C	parameter of Eq. (10)
D	diameter of sphere's with large diameter (m)
d	diameter of sphere's with small diameter (m)
d_p	sphere's diameter (m)
d_{av}	average sphere's diameter (m)
D_H	pore hydraulic mean diameter (m)
E	Pores roughness (m)
f_{Erg}	Ergun friction factor
$f/2$	Nikuradse friction factor (Eq. (20))
F_D	fraction of surface area of large particles
K_0	pores shape factor
k	permeability (m ²)
L	packing length of the bed (m)
L_{av}	average travel distance of a fluid particle (m)
n	parameter from tortuosity relation

p	parameter from tortuosity relation
Q	flow rate (m ³ /s)
R^2	correlation coefficient
Re_{Erg}	Ergun Reynolds number
Re_p	particle Reynolds number
Re_{int}	interstitial Reynolds number
r_0	sphere radius (m)
R_M	support layer resistance (m ⁻¹)
r_0/σ	packing parameter (Eq. 21))
u_D	Darcy velocity (m/s)
v	average interstitial velocity (m/s)
V_{cell}	volume of cubic cell (Eq. (21)) (m ³)
V_T	volume of pore space (m ³)
V_p	volume of spheres (m ³)
X_D	volume fraction of large spheres
$X_{D,min}$	volume fraction of large spheres in the region of minimum porosity

Greek letters

ε	porosity
ε_D^0	porosity of mono-sized packing containing large spheres
ε_d^0	porosity of mono-sized packing containing small spheres
δ	particle size ratio
λ	parameter (Eq. (12))
ΔP	pressure drop (Pa)
ζ, ζ_1, ζ_2	packing parameters (Eq. (22))
σ	characteristic dimension of the pore space available for fluid flow (m)
μ	viscosity (Pa.s)
τ	tortuosity coefficient

References

- [1] C.S. Fernandes, R.P. Dias, J.M. Nóbrega, J.M. Maia, Laminar flow in chevron-type plate heat exchangers: CFD analysis of tortuosity, shape factor and friction factor, *Chem. Eng. Process.* 46 (2007) 825–833
- [2] J. Du, X. Hu, W. Wu, B.X. Wang, A thermal dispersion model for single phase flow in porous media, *Heat Transfer Asian Res.* 32 (2003) 545–551
- [3] P.C. Carman, Fluid flow through granular beds, *Trans. Inst. Chem. Eng.* 15 (1937) 150–166.
- [4] M. Mota, J.A. Teixeira, A. Yelshin, Binary spherical particle mixed beds porosity and permeability relationship measurement, *Trans. Filtration Soc.* 1 (2001) 101–106.
- [5] P.A. Belter, E.L. Cussler, W.S. Hu, *Bioseparations – Downstream Processing for Biotechnology*, John Wiley & Sons, New York, 1988, pp. 22-23
- [6] D. Hlushkou, U. Tallarek, Transition from creeping via viscous-inertial to turbulent flow in fixed beds, *J. of Chromatogr. A*, 1126 (2006) 70-85
- [7] L. Wang, Y. Li, G. Zhao, N. Chen, Y. Xu, Experimental investigation of flow characteristics in porous media at low Reynolds numbers ($Re \rightarrow 0$) under different constant hydraulic heads, *Water* 11 (2019) 1-14
- [8] R. Dias, J.A. Teixeira, M. Mota, A. Yelshin, Tortuosity variation in a low density binary particulate bed, *Sep. Purif. Technol.* 51 (2006) 180– 184
- [9] E. Mauret, M. Renaud, Transport phenomena in multiparticle systems – I. Limits of applicability of capillary model in high voidage beds – application to fixed beds of fibers and fluidized beds of spheres, *Chem. Eng. Sci.* 52 (1997) 1807–1817
- [10] J. Comiti, M. Renaud, A new model for determining mean structure parameters of fixed beds from pressure drop measurements: Application to beds packed with parallelepipedal particles, *Chem. Eng. Sci.* 44 (1989) 1539–1545
- [11] R. Dias, J.A. Teixeira, M. Mota, A. Yelshin, Particulate binary mixtures: dependence of packing porosity on particle size ratio, *Ind. Eng. Chem. Res.* 43 (2004) 7912–7919.
- [12] S. Ergun, Fluid flow through packed columns, *Chem. Eng. Prog.* 48 (1952) 89-94.
- [13] H. Brauer, *Grundlagen der Einphasen- und Mehrphasenströmungen*, Sauerländer, Aarau & Frankfurt am Main (1971) pp. 200
- [14] K. G. Allen, T. W. Backstrom, D.G. Kroger, Packed bed pressure drop dependence on particle shape, size distribution, packing arrangement and roughness, *Powder Technol.*, 246 (2013) 590-600
- [15] R. E. Hicks, Pressure drop in packed beds of spheres, *Ind. Eng. Chem. Fundam.* 9 (1970) 500-502
- [16] I. F. Macdonald, M.S. El-Sayed, K. Mow, F.A.L. Dullien, flow through porous media – the Ergun equation revisited, *Ind. Eng. Chem. Fundam.* 18 (1979) 199-208
- [17] R. Dias, J.A. Teixeira, M. Mota, A. Yelshin, Preparation of controlled particulate mixtures with glass beads of different sizes, *Separation Purif. Technol.* 37 (2004) 69– 80.

- [18] R.P. Dias, C.S. Fernandes, M. Mota, J.A. Teixeira, A. Yelshin, Permeability and effective thermal conductivity of bisized porous media, *Int. J. Heat Mass Transfer* 50 (2007) 1295–1301
- [19] R. P. Dias, C.S. Fernandes, J.A. Teixeira, M. Mota, A. Yelshin, Permeability analysis in bisized porous media: Wall effect between particles of different size, *J. Hydrol.* 349 (2008) 470-474
- [20] M.R.J. Wyllie, A.R. Gregory, Fluid flow through unconsolidated porous aggregates: Effect of porosity and particle shape on Kozeny-Carman constants, *Ind. Eng. Chem.* 47 (1955) 1379-1388
- [21] O. Molerus, A coherent representation of pressure drop in fixed beds and of bed expansion for particulate fluidized beds, *Chem. Eng. Sci.* 35 (1980) 1331–1340
- [22] I. Iliuta, F. Larachi, B.P.A. Grandjean, Pressure drop and liquid holdup in trickle flow reactors: Improved Ergun constants and slip correlations for the Slit Model, *Ind. Eng. Chem. Res.* 37 (1998) 4542-4550
- [23] J. Tobis, Influence of bed geometry on its frictional resistance under turbulent flow conditions, *Chem. Eng. Sci.* 55 (2000) 5359-5366
- [24] A. Montillet, E. Akkari, J. Comiti, About a correlating equation for predicting pressure drops through packed beds of spheres in a large range of Reynolds numbers, *Chem. Eng. Process.* 46 (2007) 329–333