1	Experimental study of non-Darcian flow characteristics in permeable stones
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4	A manuscript prepared for Hydrology and Earth System Sciences
5	by
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21 Abstract

22 This study provides experimental evidence of Forchheimer flow and transition between 23 different flow regimes from the perspective of pore size of permeable stone. We have firstly 24 carried out the seepage experiments of permeable stones with four different mesh sizes, 25 including 24 mesh size, 46 mesh size, 60 mesh size, and 80 mesh size, which corresponding 26 to mean particle sizes (50% by weight) of 0.71 mm, 0.36 mm, 0.25 mm, and 0.18 mm. The 27 seepage experiments show that obvious deviation from Darcian flow regime is visible. In 28 addition, the critical specific discharge corresponding to the transition of flow regimes (from pre-Darcian to post-Darcian) increases with the increase of particle sizes. When the "pseudo" 29 30 hydraulic conductivity (K) (which is computed by the ratio of specific discharge and the 31 hydraulic gradient) increases with the increase of specific discharge (q), the flow regime is 32 denoted as the pre-Darcian flow. After the specific discharge increases to a certain value, the 33 "pseudo" hydraulic conductivity begins to decrease, and this regime is called the post-34 Darcian flow. In addition, we use the mercury injection experiment to measure the pore size distribution of four permeable stones with different particle sizes, and the mercury injection 35 36 curve is divided into three stages. The beginning and end segments of the mercury injection 37 curve are very gentle with relatively small slopes, while the intermediate mercury injection 38 curve is steep, indicating that the pore size in permeable stones is relatively uniform. The porosity decreases as the mean particle sizes increases, and the mean pore size can faithfully 39 40 reflect the influence of particle diameter, sorting degree and arrangement mode of porous 41 medium on seepage parameters. This study shows that the size of pores is an essential factor 42 for determining the flow regimes. In addition, the Forchheimer coefficients are also discussed in which the coefficient A (which is related to the linear term of the Forchheimer equation) is 43 linearly related to $1/d^2$ as $A = 0.0025(1/d^2) + 0.003$; while the coefficient B (which is related 44 to the quadratic term of the Forchheimer equation) is a quadratic function of 1/d as 45

46 $B = 1.14\text{E}-06(1/d)^2 - 1.26\text{E}-06(1/d)$. The porosity (*n*) can be used to reveal the effect of 47 sorting degree and arrangement on seepage coefficient. The larger porosity leads to smaller 48 coefficients *A* and *B* under the condition of the same particle size.

Keywords: permeable stone, mercury injection experiment, pore size, flow regime, non-Darcian flow.

51 1. Introduction

52 <u>Darcy (1857)</u> conducted a steady-state flow experiment in porous media and concluded 53 that the specific discharge was proportional to the hydraulic gradient, which is the Darcy's 54 law described as follow:

$$q = KJ \tag{1-1}$$

where q is the specific discharge, J is the hydraulic gradient, and K is the hydraulic conductivity. However, when the specific discharge increases above a certain threshold, deviation from Darcy's law is evident and the flow regime changes from Darcian flow regime to the so called non-Darcian flow regime (Bear, 1972), which was first observed by <u>Forchheimer (1901)</u>, who proposed a widely used non-Darcian flow equation (the Forchheimer equation) as follow:

$$J = Aq + Bq^2 \tag{1-2}$$

61 where *A* and *B* are constants related to fluid properties and pore structure. The first and 62 second terms on the right side of Eq. (1-2) roughly reflect the contributions of viscous and 63 inertial forces (or resistance to flow), respectively.

From the Forchheimer equation, we can see that when the specific discharge is sufficiently small, the inertial force can be ignored, and the equation is transformed to the form of Darcy's law. On the other hand, when the specific discharge is sufficiently large, the
viscous force can be ignored, and the equation is transformed to the fully developed turbulent
flow.

In addition to the polynomial function such as the Forchheimer equation, there are also several power-law functions proposed to describe the non-Darcian flow, and one of the most commonly used power-law equations is the Izbash equation (Izbash, 1931), which is written as:

$$J = aq^b \tag{1-3}$$

where *a* and *b* are the empirical parameters that depend on flow and materials properties, and
the coefficient *b* is usually between 1 and 2.

75 Because of its applicability for a wide range of velocity spectrum and its sound physics, 76 many scholars have adopted the Forchheimer equation (among many different types of equations) to explore the non-Darcian flow. Besides, the theoretical background of the 77 78 Forchheimer equation has been discussed in details (Panfilov and Fourar, 2006). Numerous 79 experimental data have confirmed the validity of the Forchheimer equation for a variety of 80 nonlinear flow phenomena (Geertsma, 1974; Scheidegger, 1957; Wright, 1968). The 81 quadratic Forchheimer law has also been revealed as a result of numerical modelling by 82 simulating the Navier-Stokes flow in corrugated channels (Koch and Ladd, 1996; Skjetne et 83 al., 1999; Souto and Moyne, 1997). To sum up, the Forchheimer equation will be selected as 84 a representative to describe non-Darcy flow in this study.

Since the transition between Darcian flow and non-Darcian flow is important and difficult to quantify, different scholars have carried out experiments using a wide range of porous media, including homogeneous and heterogeneous porous media. Most of the experimental studies have focused on the influence of mean particle size on flow state

89 transition using homogeneous porous media. In fact, it was believed that the nonlinear (or 90 non-Darcian) flow behavior in porous media was due to turbulent effect of flow in earlier 91 studies and the Reynold number (Re) was widely used to quantify the initiation of non-92 Darcian flow. Bear (1972) concluded that the critical Re (denoted as Re_c) of flow states (or 93 the Re value at which flow starts to change from Darcian flow regime to non-Darcian flow 94 regime) is between 1 to 10. This finding was based on experimental data collected in packed 95 sand beds (Ergun, 1952; Fancher and Lewis, 1933; Lindquist, 1933; Scheidegger, 1960). Schneebeli (1955) and Wright (1968) experimentally measured the value of Re at the 96 97 beginning of turbulence and concluded that at very high velocities, the deviation from 98 Darcy's law is due to inertial effects followed by turbulent effects. In addition, Dudgeon 99 (1966) confirmed that Re_c is about 60~150 for relatively coarse particle medium including 100 river gravels, crushed rock particles and glass marbles with grain sizes from 16 mm to 152 101 mm. Dudgeon (1966) indicated that the deviation from Darcy's law was not entirely due to 102 turbulence, but in a large extent due to inertial forces. Besides, Geertsma (1974) proposed an 103 empirical relationship among the inertial coefficient, permeability and porosity by conducting 104 non-Darcian flow experiments in unconsolidated and consolidated sands. The laser 105 anemometry and flow visualization studies of fluid flow in porous structures were used by Dybbs and Edwards (1984), and they observed the nonlinear behavior at Reynolds numbers 106 107 around 150. Latifi et al. (1989) found that the transition from unsteady-state laminar flow to 108 non-Darcian flow in packed beds of spheres was between Re values of 110 and 370. Seguin et 109 al. (1998) investigated the characterization of flow regimes in various porous media with 110 electrochemical techniques and found that the end of the Darcian flow regime in packed beds 111 of particles appeared at Re about 180. Besides, Bu et al. (2014) indicated that the Darcian 112 flow in the packed beds would end at *Re* around 100 by using electrochemical techniques. 113 Sedghi-Asl et al. (2014) found that the Darcy's law was usually not valid for rounded particle 114 sizes greater than 2.8 mm, according to the experimental results of flow in different sizes of rounded aggregates. Our previous experimental research (Li et al., 2017) indicated that when 115 the particle size was smaller than 2.8 mm, the flow state gradually changed from pre-Darcy 116 117 flow to post-Darcy flow when the specific discharge increased. When the medium particle 118 sizes get even larger, such as 4.5 mm, 6.39 mm, 12.84 mm, and 16 mm (Moutsopoulos et al., 119 2009), only the post-Darcy flow exists. Based on above analysis, we can see that many 120 previous experiments were carried out on homogeneous porous media, and the non-Darcy 121 flow characteristics are quite different in porous media with various particle sizes.

122 Among the numerous experimental studies on this issue, it is evident that most of them 123 focused on the effect of the mean particle size rather than the particle size distribution. 124 Recently, a few investigators recognized the importance of particle size heterogeneity in 125 understanding the transition of flow regimes, and have carried out a series of experiments to 126 address the issue. For instance, Van Lopik et al. (2017) provided new experimental data on 127 nonlinear flow behavior in various uniformly graded granular material for 20 samples, 128 ranging from medium sands ($d_{50} > 0.39$ mm) to gravel ($d_{50} > 6.34$ mm). In addition, they 129 investigated the nonlinear flow behavior through packed beds of 5 different types of natural 130 sand and gravel from unconsolidated aquifers, as well as 13 different composite mixtures of uniformly graded filter sands at different grain size distributions and porosity values (Van 131 132 Lopik et al., 2019). We have also discussed the effect of particle size distribution on 133 Forchheimer flow and transition of flow regimes in a previous study (Li et al., 2019b). And 134 our study showed that the uniformity coefficient of porous media (a term used to describe the 135 pore size distribution) is a critical factor for determining the flow regimes besides the mean 136 particle sizes. Yang et al. (2019) investigated the effects of the particle size distribution on the 137 seepage behavior of a sand particle mixture subjected and evaluated the validity of empirical 138 formulas of permeability and inertia factor used in engineering practice. Shi et al. (2020)

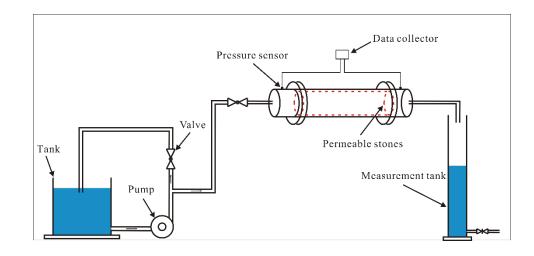
discussed the non-Darcy flow behavior of granular limestone with a wide range of porosity from 0.242 to 0.449. Based on the experimental data, <u>Shi et al. (2020)</u> proposed an empirical hydraulic conductivity-porosity relation as well as an expression of inertial coefficient. Regardless of the media investigated are homogeneous or heterogeneous, the essence of the water passing capacity of porous media is pore sizes. Thus, exploring the distribution of pores in porous media is the basis of studying flow dynamics of Darcian and non-Darcian flows.

145 The purpose of this study is to provide a quantitative analysis on the effects of pore size on the transition of flow regimes between Darcian and non-Darcian flows based on a series of 146 147 laboratory experiments. To meet the objectives, we have firstly carried out the seepage 148 experiments of permeable stones with four different particle sizes. After that, we have 149 conducted mercury injection experiments on permeable stones with four different particle 150 sizes, and the pore size distributions with different particle sizes are obtained. Finally, the 151 effect of pore size on the transition of flow regimes and Forchheimer coefficients are discussed based on the experimental results. 152

153 **2. Experimental methodology**

154 **2.1 Experimental setup and methods**

155 The experimental device is mainly composed of three parts: a water supply device, a 156 seepage experimental device and a measuring device. The schematic diagram of the 157 experimental apparatus is shown in Fig. 1. The water supply device consists of a tank, a centrifugal pump and a flow regulating valve. The seepage experimental device consists of a 158 159 permeable stone and a plexiglass column. The measurement device monitors the real-time 160 water temperature and pressure. The water temperature is measured using a thermometer with 161 a precision of measurement of 0.1 °C. The water-level fluctuation is measured to calculate the 162 flow rate by a pressure transducer (CY201, Chengdu test LLC, China) in the range of 0–20 163 kPa with ±0.1% accuracy. The measuring device consists of a cylindrical tank and a pressure transducer. The sample of permeable stone is 60 mm in length with a circular cross section of 51.3 mm in diameter. Two pressure transducers are set at the entrance and exit of the column to measure the pressure drop. To minimize the boundary effects, the pressure transducer is placed 30 mm away from either end of the column, and the way of pressure measurement is consistent with our previous studies (Li et al., 2017; Li et al., 2019b).





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Fig. 1 The schematic diagram of experimental apparatus.

171 **2.2 Experimental Materials and Procedures**

172 Four different particle sizes of permeable stones are selected to carry out the seepage experiment in this study. It is necessary to make a brief overview of the preparation process 173 of permeable stone, which is a type of artificially made tight porous medium formed by sand 174 175 grains and cementing compound. In the process of preparing permeable stone, a certain 176 particle size of sand and cementing compound is put in a mold, and is consolidated at room 177 temperature. Permeable stone is widely used in urban road design, sponge city construction 178 and ecological effect research (Guan et al., 2021; Li et al., 2019a; Yu et al., 2021). And the 179 most commonly used permeable base materials are large pore cement stabilized macadam, large diameter permeable asphalt mixture and so on (Suo et al., 2021). For permeable stone, 180 181 there must be a certain connected pore space to maintain a certain permeability for 182 transmitting water. However, the increase of pore space will lead to the decrease of pavement

performance and mechanical strength (Han et al., 2016; Wang et al., 2021). Therefore, many 183 184 scholars have carried out a lot of research on controlling the proper pore space of permeable stone (Alvarez et al., 2010; Prowell et al., 2002; Xie and Watson, 2004). We have carried out 185 186 the seepage experiments of permeable stones with four different mesh sizes, including 24 mesh size, 46 mesh size, 60 mesh size, and 80 mesh size, and the mesh size is defined as the 187 188 number of mesh elements (all in square shapes) in a one inch by one inch square, thus a 189 greater number of mesh size implies a smaller particle size. For instance, we can convert 190 above four different mesh sizes of permeable stones into corresponding particle sizes of 0.71 191 mm, 0.36 mm, 0.25 mm and 0.18 mm, respectively. The pore structure of permeable rock will 192 not change in the process of the seepage experiment under room temperature, and the 193 physical diagrams of four kinds of permeable stones with different particle sizes are shown in 194 Fig. 2 and Fig. 3.



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Fig. 2 Physical drawing of permeable stones with four different particle sizes.



(b)

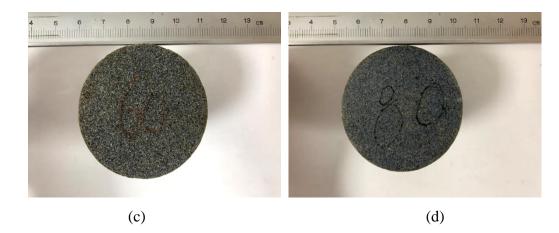


Fig. 3 Permeable stones with different particle sizes: (a) 24 mesh size or 0.71 mm, (b) 46 mesh size or 0.36 mm, (c) 60 mesh size or 0.25 mm, and (d) 80 mesh size or 0.18 mm.

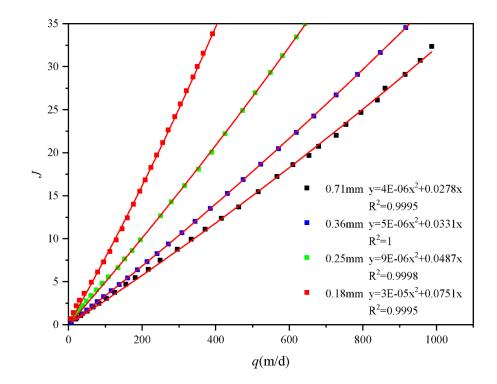
203 It is worth mentioning that the contact surface of the sample and the plexiglass column 204 is sealed to prevent any preferential flow through the wall of the plexiglass column. After the 205 permeable stone is inserted into the plexiglass column, both ends are sealed with silicone glue. 206 The water passing through the permeable stone is then collected by a cylindrical tank. Moreover, the ratio of the internal diameter of the column to the particle size of permeable 207 stone is greater than 12, which can eliminate any possible wall effect on the seepage 208 209 according to **Beavers et al.** (1972). When carrying out the experiment, it usually takes about 210 two hours to saturate the permeable stone. For each packed sample, more than 25 tests with 211 different constant inlet pressures were conducted under steady-state flow condition. In 212 addition, for each group of permeable stone, repeated tests under the same experimental 213 condition were carried out 3-4 times to ensure the accuracy of the results.

214 **3. Results and discussion**

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215 **3.1 Permeable stone seepage experiment**

In this study, we selected permeable stone with four different particle sizes as the research objects, including 24 mesh size, 46 mesh size, 60 mesh size and 80 mesh size. The mesh size is the number of holes per inch of screen mesh and the particle size is inversely 219 proportional to the mesh size. The mean particle sizes corresponding to the four different 220 mesh sizes are 0.71 mm, 0.36 mm, 0.25 mm, and 0.18 mm, respectively, where the mean 221 particle size is corresponding to 50% by weight hereinafter in this study. Such a definition of 222 mean particle size may be different from some other studies such as Fetter (2001) which has 223 used 10% by weight as the mean particle size. The relationship between the specific 224 discharge (q) and the hydraulic gradient (J) of permeable stones is plotted in Fig. 4. The units 225 of specific discharge mentioned in this study are all converted to meters per day (m/d). To 226 better compare with the actual groundwater flow, we converted the specific discharge to 227 meters per day (m/d). Therefore, the best-fitting exercise yields Forchheimer numbers with 228 orders of magnitudes to be about -4. In addition, the critical Forchheimer numbers proposed 229 by Zeng and Grigg (2006) and Javadi et al. (2014) are empirical, in fact, the transition 230 between Darcy to non-Darcy is successional over a certain range of Forchheimer numbers. 231 The non-Darcian flow criterion applicable to different pore media is established by 232 conducting seepage resistance experiments in homogeneous and heterogeneous porous media 233 in our previous study (Li et al., 2017; Li et al., 2019b), which is consistent with the results of 234 Zeng and Grigg (2006). Generally speaking, the q-J and q-K curves are the most commonly 235 used methods to analyze flow regime when conducting seepage resistance experiments in 236 porous media. However, the nonlinear characteristics of q-J curve are not obvious due to the 237 relatively small velocity range used in the experiments. The traditional hydraulic conductivity 238 is the ratio of the specific discharge versus the hydraulic gradient (q/J), and it is a constant if 239 Darcy's law is applicable, which is denoted as K_D (Li et al., 2019b). In fact, the ratio of q/J is 240 no longer a constant for the problems discussed in this study. In a word, the *q*-*K* curve can be 241 used to observe the transition of flow state more intuitively.

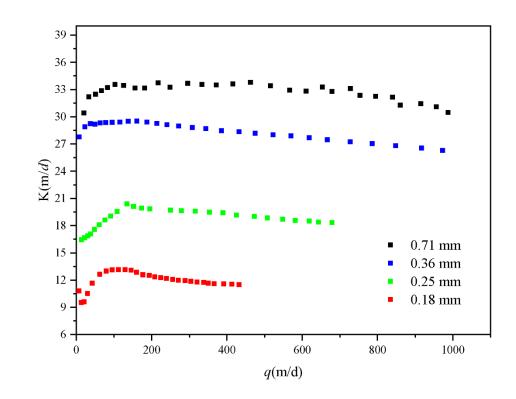


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Fig. 4 Variation of J with q of four permeable stones with different particle sizes.

244 Fig. 4 shows that when q is somewhat the same, a larger mesh size (which means a 245 smaller particle size) will lead to a larger J. And the results are consistent with our previous 246 studies (Huang et al., 2013; Li et al., 2017; Li et al., 2019b). However, the nonlinear 247 characteristics of q-J curve are not obvious due to the relatively small velocity range used in 248 the experiments. Nevertheless, the best-fitting results using the Forchheimer equation are 249 satisfactory. To analyze the influence of pore size on seepage flow regimes, we have obtained the relationship between q and the "pseudo" hydraulic conductivity (K) (which is computed 250 251 using q/J) of four permeable stones with different particle sizes, as shown in Fig. 5. We 252 should point out that the "pseudo" hydraulic conductivity term discussed here for non-Darcian flow is usually not a constant, thus it is different from the hydraulic conductivity 253 254 term used in Darcy's law, which is a constant. It is obvious that the hydraulic conductivity is 255 not a constant with the increase of specific discharge, so it is called the "pseudo" hydraulic conductivity (Li et al., 2019b). 256



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Fig. 5 Variation of *K* with *q* of four permeable stones with different particle sizes. We can divide the *q*-*K* curve into two segments: for the first segment, *K* increases with the increase of *q*, which is denoted as the pre-Darcian flow. For the second segment, after *q* increases to a certain value, *K* begins to decrease with *q*, which is called the post-Darcian flow. In fact, Izbash (1931) presented the equation as $q = M \left(\frac{dH}{dx}\right)^m = Mi^m$, where *M* and *m*

263 are the coefficients determined by fluid flow and properties of porous media. When m=1, the 264 Izbash equation reduces to Darcy law, when m>1, the Izbash equation corresponds to the pre-265 Darcy flow and when m < 1, the Izbash equation refers to the post-Darcy flow (Dejam et al., 2017; Soni et al., 1978). Besides, Dejam et al. (2017) carried out a more detailed study on 266 267 issues related to the pre-Darcy and post-Darcy flows. And the influence of pre-Darcy flow on 268 the pressure diffusion for homogenous porous media is studied in terms of the nonlinear exponent and the threshold pressure gradient. When the hydraulic gradient is small (and q is 269 270 small as well), a great portion of water is bounded (or becomes immobile) on the surface of 271 solids due to the solid-liquid interfacial force, and only a small fraction of the water is mobile

272 and free to flow through the pores. In addition, another justification for the pre-Darcy 273 behavior may be due to an effect of a stream potential which generates small countercurrents 274 along pore walls in a direction opposite that of the main flow (Bear, 1972; Scheidegger, 2020). 275 And Swartzendruber (1962b) stated that the surface forces arose in a solid-fluid interface due 276 to strong negative charges on clay particle surfaces and the dipolar nature of water molecules 277 caused a pressure gradient response to be nonlinear and led to the pre-Darcy flow 278 (Swartzendruber, 1962a). As the hydraulic gradient increases (and q increases as well), the 279 initial threshold for mobilizing the previously immobile water near the solid-liquid surface is 280 overcome and more water participates in the flow. For this reason, the "pseudo" hydraulic 281 conductivity increases with the increase of hydraulic gradient and the specific discharge in 282 the first segment. When the specific discharge increases to the critical specific discharge (q_c) , the "pseudo" hydraulic conductivity is maximized. According to $K = \frac{q}{Aq + Bq^2} = \frac{1}{A + Bq}$ 283 based on Eq. (1-2), we can find that the "pseudo" hydraulic conductivity begins to decrease 284

as the specific discharge continues to increase. Besides, the critical specific discharge corresponding to the transition of flow regimes (from pre-Darcian to post-Darcian) increases with the increase of particle sizes (or decrease of mesh sizes).

288 **3.2 Mercury injection experiment**

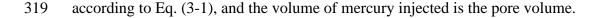
The particle size, different grain size distributions and degree of sorting are the main factors that determine the size and shape of pores. And the shape of the pores determines the tortuosity and distribution of flow paths, which are related to viscous and inertial flow resistances. It is generally accepted in previous studies that the pore sizes of porous media have an impact on the seepage law (Maalal et al., 2021; Zhou et al., 2019). However, the structure of natural porous media is very complex, and it is difficult to quantify the effects of the arrangement of particles on the seepage law. The characteristics of pore size distribution 296 contains critical information for quantifying the flow regimes. The mercury intrusion 297 porosimetry and the nitrogen adsorption isotherm are two commonly used methods to characterize the pore sizes and their distribution (Rijfkogel et al., 2019). Besides, other 298 299 techniques can also be used to derive the pore size distribution, such as small-angle neutron 300 and X-ray scattering measurements, CT images and nuclear magnetic resonance (Anovitz and 301 Cole, 2015; Hall et al., 1986; Kate and Gokhale, 2006; Lindquist et al., 2000). In this study 302 we will use the mercury injection experiment to measure the pore size distribution of the four 303 permeable stones with different particle sizes and use the information to describe the flow 304 regimes.

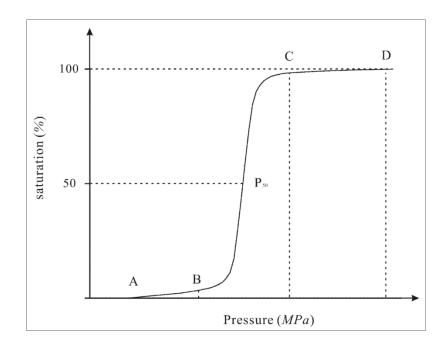
To quantitatively study the pore size and pore throat distribution, we need to envisage a physically based conceptual model to describe the pore structures of permeable stones. The commonly used model is the so-called capillary model (Pittman, 1992; Rezaee et al., 2012; Schmitt et al., 2013), which approximates the connected pores as many paralleled capillaries. And the capillary forces are generated at the phase interface due to the surface tension between the solid and liquid phases when liquid flows in a capillary. The capillary force is directed toward the concave liquid level, and is shown as (Washburn, 1921):

$$P_c = \frac{2\sigma\cos\theta}{r} \tag{3-1}$$

312 where P_c is the capillary force, σ is the solid-liquid interfacial tension, θ is the wet angle 313 between the liquid and the solid surface, and *r* is the radius of curvature in capillary.

Since mercury is a nonwetting phase to solids, so to get mercury into the pores of the permeable stone, an external force (or displacement pressure) must be applied to overcome the capillary force. When a greater pressure is applied, mercury can enter smaller pores. When a certain pressure is applied, the injection pressure is equivalent to the capillary pressure in the corresponding pore. Then we can calculate the corresponding capillary radius





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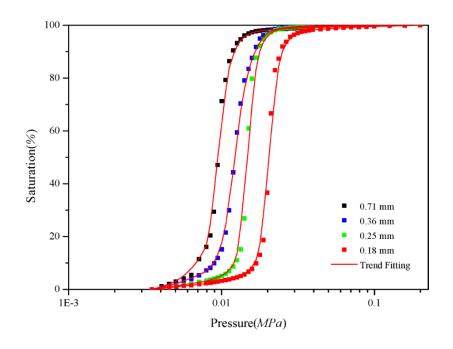
Fig. 6 Schematic diagram of pressure changes with saturation: the initial stage (A-B), the intermediate mercury entry stage (B-C), and the end stage (C-D).

By continuously increasing the injection pressure, one can obtain the curve of injection pressure and the volume of injected mercury, from which one can also obtain the pore-throat distribution curve and capillary pressure curve. According to the amount of mercury injected at different injection pressures, the relation between the injection pressure and the injection saturation is shown in Fig. 6.

328 Fig. 6 shows that the mercury injection curve can be divided into three stages. Firstly, 329 during the initial stage (A-B) which has a very mild slope, the intake pressure is very small 330 and the intake saturation is also very low. With the increasing of the injection pressure, the 331 intake saturation slowly increases. Secondly, during the intermediate mercury entry stage (B-332 C) which has a steep slope, a small pressure change will lead to a significant saturation 333 change. This means that the pores are relatively uniform and the differences in pore sizes are 334 small. It is well known that for mercury injection experiments, as injection pressure increases, 335 the injection saturation will gradually increase and eventually all the pores will be filled with

336 mercury. As can be seen from Fig. 7, with the continuous injection of mercury, the pressure of 337 permeable stones with different particle sizes varies with saturation, which is reflected in the 338 different pressure P_B and P_C at different stages. However, the reason for the different pressure 339 is the difference of pore size distribution in the permeable stones. Therefore, the pressure 340 ratio of B and C (P_C/P_B) can be used as one of the criteria to characterize the heterogeneity of 341 pore size in porous media. Besides, when the saturation reaches 50%, the corresponding 342 pressure value (P_{50}) reflects the characteristics of the mean pore size, and a larger P_{50} leads to a larger mean pore size. Finally, during the end stage (C-D) which has a very mild slope as 343 344 well, the amount of mercury will not increase considerably when the injection pressure 345 increases. This indicates that nearly all the pores are essentially filled with mercury, and the 346 mercury injection experiment is completed. After completing the mercury injection 347 experiments, we have obtained the mercury injection curves of four permeable stones with 348 different particle sizes, as shown in Fig. 7.

349 We can make a number of interesting observations based on Fig. 7. Firstly, the pressure 350 at the starting point (when the saturation begins to increase), denoted as $P_{\rm A}$, increases as the 351 mean particle size decreases. This means that the maximum pore size in permeable stone 352 decreases with the decrease of the mean particle sizes. Secondly, the mercury injection curves 353 of four permeable stones all include steep intermediate stages, indicating that the pore size 354 distributions are all relatively uniform. And the corresponding pressure values at points B and 355 C increase as the mean particle sizes decreases. Moreover, the pressure ratios corresponding 356 to points B and C (P_C/P_B) also decrease with the decrease of particle sizes, suggesting even 357 more uniform pore size distributions with decreasing particle sizes. Thirdly, the intermediate 358 mercury entry stages gradually shift to the right with the decrease of particle sizes. When the 359 saturation reaches 50%, the corresponding pressure (the median pressure) decreases with the 360 increase of the mean particle sizes. Fourthly, the mercury injection curves of these four 361 permeable stones with different particle sizes all approach 100% saturation with very mild 362 slopes, indicating that there are few small pores in the permeable stones. We have extracted 363 the key pressure characteristic values of mercury injection experiment of Fig. 7, and listed the 364 results in Table 1.



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366 Fig. 7 Variation of pressure with saturation of four permeable stones with different particle

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sizes.

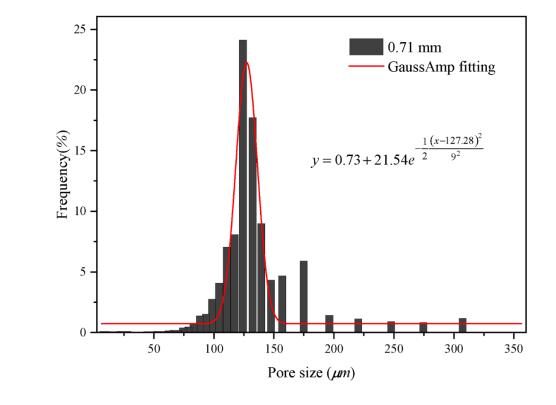
368 Table 1. Pressure characteristic values of four permeable stones with different particle sizes.

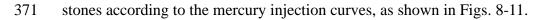
Mesh size	$P_{\rm A}(MPa)$	$P_{\rm B}(MPa)$	$P_{\rm C}(MPa)$	$P_{50}(MPa)$	$P_{\rm C}/P_{\rm B}$
24	0.0041	0.0064	0.0133	0.0094	2.0987
46	0.0045	0.0071	0.0188	0.0119	2.6374
60	0.0051	0.0112	0.0211	0.0150	1.8764
80	0.0057	0.0158	0.0281	0.0211	1.7758

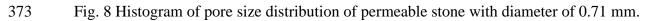
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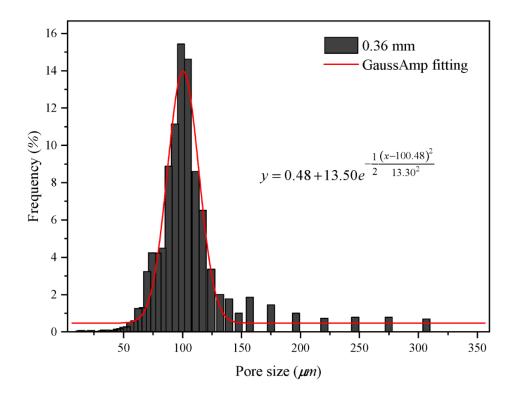
To observe the pore size distributions of the four permeable stones with different particle

370 sizes in more details, we can calculate the percentages of different pore sizes in permeable









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Fig. 9 Histogram of pore size distribution of permeable stone with diameter of 0.36 mm.

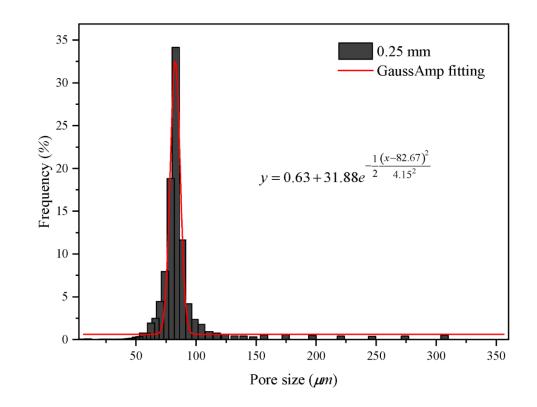
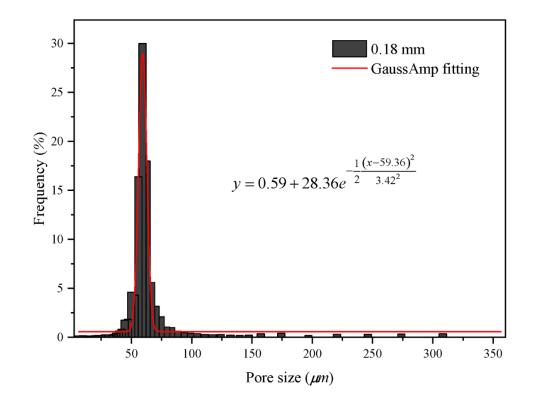
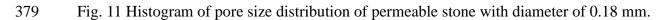


Fig. 10 Histogram of pore size distribution of permeable stone with diameter of 0.25 mm.





From Fig. 8 to Fig. 11 we can find that the pore sizes of the four permeable stones are

uniform and fall within narrow ranges. The pore size distributions of four different particle sizes show a skewed normal distribution. Besides, the pore maximum proportion (the peak of the curve, see Figs. 8-11) of permeable stones with different particle sizes are different, which are $124 \ \mu m$, $99 \ \mu m$, $83 \ \mu m$ and $59 \ \mu m$, respectively. The Gaussian function is widely used to characterize the pore system and classify the petrophysical rock (Harlan et al., 1995; Jeon et al., 2014; Xu and Torres-Verdín, 2013), and the general form of the Gauss function is shown below:

$$y = y_0 + He^{\frac{(x-x_c)^2}{2w^2}}$$
(3-2)

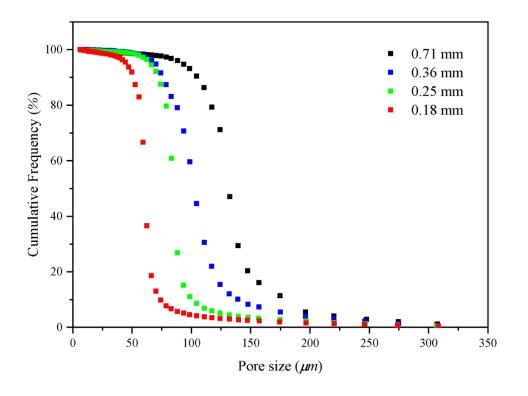
where H is the height of the peak of the mercury injection curve, x_c is the abscissa 388 389 corresponding to the peak of the curve (the pore size), w is the standard variance, which 390 represents the width of the curve. To characterize the distribution of pore structure of four 391 different permeable stones, we best-fit the Gaussian curve of the pore distribution of four 392 permeable stones with different particle sizes, and the best-fitted parameters are shown in 393 Table 2. We can make several interesting observations from Table 2. Firstly, the expected value (x_c) decreases with decreasing particle sizes of permeable stone, and the x_c values of 394 395 different permeable stones are almost the same. Secondly, the standard variance (w) 396 corresponding to the permeable stone of 0.18 mm is the smallest, indicating that the pore size 397 distribution is more concentrated (or relatively homogeneous). For comparison, the pore size 398 distribution of 0.36 mm permeable stone is the widest with the greatest variance. Finally, different values of H represent different proportions of pore sizes, among which the highest 399 proportion can reach 34.04%. It will be desirable to establish a correlation between the 400 401 parameters used in the pore-size distribution of Eq. (3-2) with the two Forchheimer 402 coefficients A and B. This objective may be achieved using high-resolution pore-scale fluid 403 mechanics simulations, which are out of the scope of this study. Further research is needed to

404 address this issue in the future.

Mesh size	particle size (mm)	<i>y</i> 0	Н	X_C	W
24	0.71	0.73	21.54	127.28	9.00
46	0.36	0.48	13.49	100.48	13.30
60	0.25	0.63	31.88	82.67	4.15
80	0.18	0.59	28.36	59.36	3.42

405 Table 2. Gaussian function characteristic values of four permeable stones with different406 particle sizes.

407 The pore size distributions fall within ever narrower ranges with mesh sizes become 408 larger. Moreover, the cumulative percentage frequency curves of the pore size distributions 409 with different particle sizes are exhibited in Fig. 12 and the results are shown in Table 3.





411

Fig. 12. The cumulative frequency curve of pore size distribution.

412	Fig. 12 shows that R_{50} (the pore size corresponding to the median pressure P_{50}) increases
413	with the increase of permeable stone particle size, and the mean pore diameter (R_m) also
414	increases. In general, the pore size corresponding to the median pressure (denoted as R_{50})
415	may be slightly different than the mean pore diameter (R_m) which has been defined in
416	different ways by various investigators when analyzing the pore size distributions (Hea and
417	Zhangb, 2015; Zhen-Hua et al., 2007; Zhihong et al., 2000). As R ₅₀ is easily identifiable in
418	the mercury injection experiments, it is used in this study as a representative of the mean pore
419	diameter (R_m) of the permeable stone. Besides, the seepage law of permeable stone is closely
420	related to the pore size, and the smaller average pore size will result in a larger hydraulic
421	gradient under the condition of the same specific discharge (see Fig. 4). The pore size
422	characteristic values with different particle sizes are listed in Table 3. We find that the
423	porosity decreases as the particle size increases while the mean pore diameter increases. And
424	the mean pore size can reflect the influence of particle diameter, sorting degree and
425	arrangement mode of porous medium on seepage parameters.

426 Table 3. Pore size characteristic values of four permeable stones with different particle size
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Mesh size	Mean particle size (mm)	Porosity (%)	$R_m\left(\mu m\right)$	R ₅₀ (µm)
24	0.71	32.35	131.31	131.34
46	0.36	36.69	102.56	103.42
60	0.25	40.82	84.73	85.09
80	0.18	42.88	60.97	61.12

Note: R_m is the mean pore diameter, R_{50} is the pore diameter corresponding to the median 428 pressure P_{50} .

429 **3.3** Analysis of influencing factors of Forchheimer equation coefficients

430 **3.3.1 Influence of particle size on equation coefficient**

The analysis of non-Darcy coefficient has always been of interest to many researchers working in different disciplines of porous media flow (Moutsopoulos et al., 2009; Sedghi-Asl et al., 2014; Shi et al., 2020). Various studies have suggested expressions for Forchheimer coefficients, different scholars obtained numerous datasets through different experiments and simulation methods to quadratic best-fitting the specific discharge-hydraulic gradient curves. And the coefficients of different fitting equations are shown in the following Table 4.

Equations	Coefficient $A(s/m)$	Coefficient $B(s^2/m^2)$
Ward (1964)	$A = \frac{360}{gd^2}$	$B = \frac{10.44}{gd}$
<u>Blick (1966)</u>	$A = \frac{32}{gnd^2}$	$B = \frac{C_D}{2gn^2d}$
<u>Ergun (1952)</u>	$A = \frac{150(1-n)^2}{gn^3d^2}$	$B = \frac{1.75(1-n)}{gn^3d}$
Macdonald et al. (1979)	$A = \frac{180(1-n)^2}{gn^{3.6}d^2}$	$B = \frac{1.8(1-n)}{gn^{3.6}d}$
<u>Kovács (1981)</u>	$A = \frac{144(1-n)^2}{gn^3d^2}$	$B = \frac{2.4(1-n)}{gn^3d}$
Kadlec and Knight (1996)	$A = \frac{255(1-n)^2}{gn^{3.7}d^2}$	$B = \frac{2(1-n)}{gn^3d}$
<u>Irmay (1964)</u>	$A = \frac{180(1-n)^2}{gn^3 d^2}$	$B = \frac{0.6(1-n)}{gn^3d}$

437 **Table 4.** The Forchheimer coefficients of empirical relations.

438 <u>Sidiropoulou et al. (2007)</u> focused on the determination of the Forchheimer coefficients 439 for non-Darcian flow in porous media and evaluated the original theoretical equations above 440 and the validity of these equations was checked using existing experimental data. In addition, 441 the Root Mean Square Error (RMSE) was used as a criterion to quantitatively evaluate the 442 coefficients, and the RMSE was defined as $RMES = \sqrt{\frac{\sum_{i=1}^{N} (x_i - y_i)^2}{N}}$, where x_i were the 443 experimental values of Forchheimer coefficients, y_i were the values computed by different 444 equations above, and N was the total number of experimental points (Moutsopoulos et al., 445 2009). The different forms of Forchheimer coefficients described above are based on different 446 assumptions and simplifications of pore structure. Consequently, these series of coefficients 447 are applicable under specific conditions with different degrees of accuracy.

448 According to Eq. (1-2), the hydraulic gradient (*J*) is composed of a viscous force-related 449 component (J_n) and an inertia force-related component (J_r), and for specific derivation, please 450 refer to previous studies (Huang, 2012):

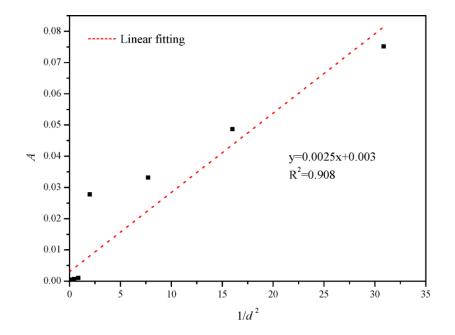
$$J_n = Aq = \frac{\alpha\mu}{\rho g} \frac{1}{d^2} q \quad J_r = \frac{\beta}{g} \frac{1}{d} q^2$$
(3-3)

451 We can see from Eq. (3-3) that the J_n is inversely proportional to the square of the particle 452 size, and the J_r is inversely proportional to the particle size when the specific discharge 453 remains the same. Both J_n and J_r are closely related to specific surface area and sizes of pores. 454 Therefore, the particle size is an important factor affecting the Forchheimer coefficient, 455 Huang et al. (2013) carried out the experimental investigation on water flow in four columns 456 with cubic arrays of acrylic balls in diameter 3 mm, 5 mm, 8 mm and 10 mm, where all the acrylic balls are arranged in regular cubes. Accordingly, the coefficients A and B can be 457 458 written as follows:

$$A = \frac{\alpha \mu}{\rho g} \frac{1}{d^2} \qquad B = \frac{\beta}{g} \frac{1}{d}$$
(3-4)

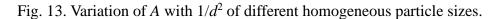
459 where α and β are constants related to the shape, sorting, and arrangement of the particles, 460 and the specific derivation process is detailed in the previous study (<u>Huang, 2012</u>). The 461 experimental results showed that the coefficient *A* was inversely proportional to the particle 462 diameter square (d^2) and coefficient *B* was inversely proportional to the particle size (d)463 (Huang et al., 2013).

464 The uniform diameter cubic arrangement of porous media mentioned above is a rather ideal medium. The shape and arrangement of particles of natural pore aquifers are usually 465 irregular. Therefore, the above-mentioned linear correlations between A and $1/d^2$, and 466 between B and 1/d should be examined specifically. For this purpose, we collect the 467 experimental data of homogeneous porous media, including the previous research results and 468 469 the results of other scholars. Among them, samples P1-P4 are the permeable stones selected 470 in this study, samples L1-L5 are from previous studies (Li et al., 2017), and the experimental 471 data of samples M1-M4 are from Moutsopoulos et al. (2009). The fitting coefficients are 472 shown in Table 5. Furthermore, we can identify nice correlations between the Forchheimer coefficient A and $1/d^2$ and between the Forchheimer coefficient B and 1/d, which are shown 473 474 in Fig. 13 and Fig. 14, respectively.

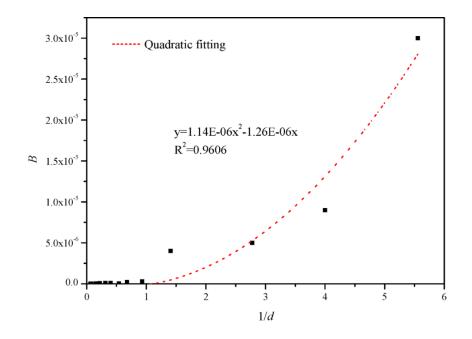


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Fig. 14. Variation of *B* with 1/d of different homogeneous particle sizes.

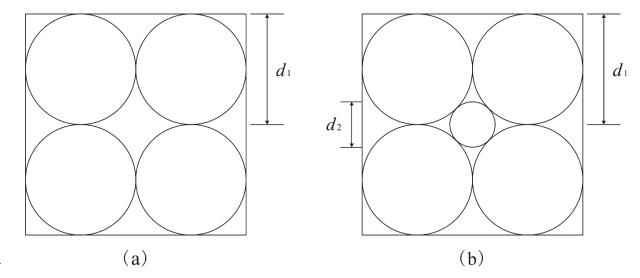
We can see from Fig. 13 that the coefficient A is linearly related to $1/d^2$ and the 479 relationship between coefficient A and is given as $A = 0.0025(1/d^2) + 0.003$. And the 480 relationship between coefficient B and 1/d is completely different from the linear correlation 481 482 as reported before. Fig. 14 shows that the coefficient B is quadratic related to 1/d and the relationship between coefficient B and 1/d is given as $B = 1.14\text{E}-06(1/d)^2 - 1.26\text{E}-06(1/d)$. 483 That is to say, the relationship between coefficient A and $1/d^2$ is consistent with the law of 484 485 simple cubic arrangement porous media, but the relationship between coefficient B and 1/d is not consistent with the law of simple cubic arrangement porous media. The structure of 486 487 porous medium arranged in cubes is different from the permeable stone. The porosity of the 488 porous media with spheres arranged in cubic is close to 0.48, independent of the diameter of 489 spheres. While the particle shape, arrangement and tightness of permeable stone are different, 490 and the porosity of permeable stone with different particle size is also different (see Table 3).

491 Table 5. Experimental fitting coefficient of different homogeneous particle sizes.

Sample	Particle size (mm)	Fitting equation	A	В	The correlation
P1	0.18	y=0.0751x+3E-05x ²	0.0751	3E-05	0.9995
P2	0.25	y=0.0487x+9E-06x ²	0.0487	9E-06	0.9998
P3	0.36	y=0.0331x+5E-06x ²	0.0331	5E-06	1
P4	0.71	y=0.0278x+4E-06x ²	0.0278	4E-06	0.9995
L1	1.075	y=0.001x+3E-07x ²	0.001	3E-07	0.9999
L2	1.475	y=0.0007x+2E-07x ²	0.0007	2E-07	0.9998
L3	1.85	y=0.0005x+5E-08x ²	0.0005	5E-08	0.9998
L4	2.5	y=0.0005x+9E-08x ²	0.0005	9E-08	0.9997
L5	3.17	y=0.0004x+1E-07x ²	0.0004	1E-07	0.9998
M1	4.5	y=3E-05x+7E-08x ²	3E-05	7E-08	0.9913
M2	6.39	y=3E-05x+3E-08x ²	3E-05	3E-08	0.9984
M3	12.84	y=1E-05x+2E-08x ²	1E-05	2E-08	0.9977
M4	16	y=1E-05x+2E-08x ²	1E-05	2E-08	0.998

492 **3.3.2 Influence of porosity on equation coefficient**

In above sections, we have analyzed the influence of particle sizes on seepage coefficient. Furthermore, the pore size and pore specific surface area are also related to the arrangement and sorting degree of particles, that is, to the porosity of porous media. To explore the effect of sorting degree on seepage coefficient, we draw a schematic diagram of different sorting degree of particles, as shown in Fig. 15. The degree of particle sorting is one of the important factors affecting the pore size. In porous media with a poor sorting degree,
the pore size is usually determined by the diameter of the smallest particle. We can see from
Fig. 15 that the pores between the larger particles are filled by smaller particles, resulting in
even smaller pores. In addition, the poorer sorting degree of particles leads to the larger pore
specific surface area and stronger viscous force of flow, which can lead to a larger coefficient *A*.





505

Fig. 15. The schematic diagram of different particle sorting with cube arrangement.

506 Furthermore, we have also provided the schematic diagrams of spherical particles with equal size in two simple arrangements, namely cube arrangement and hexahedron 507 508 arrangement, as shown in Fig. 16. And the cube arrangement is the less compact arrangement 509 with a pore diameter of 0.414*d*, while the hexahedron arrangement is the more compact 510 arrangement with a pore diameter of 0.155d. The characteristic value of pore structure in 511 different arrangement with the same particle size are shown in Table 6. We can see that 512 different arrangement modes will substantially affect the pore specific surface area and pore 513 size of porous media. The more compactly packed particles lead to the larger pore specific 514 surface area and stronger viscous force. Meanwhile, the smaller pore diameter is associated 515 with stronger effect of viscous force and inertia force. In summary, the better sorting degree of particles leads to the weaker viscous and inertial forces, then the coefficients A and B will be smaller. As the better sorting degree and the less compact (or looser) arrangement particles mean the larger porosity, so we can conclude that the larger porosity leads to the smaller coefficients A and B under the condition of the same particle size.

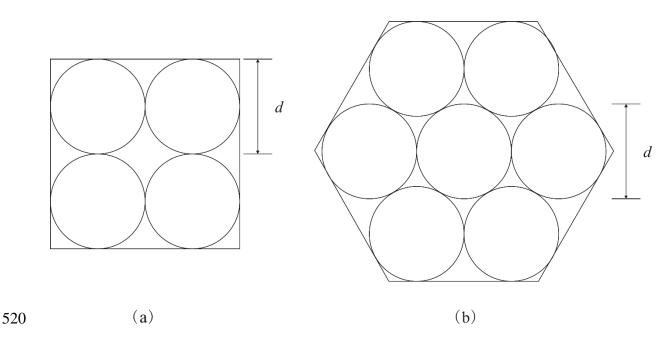
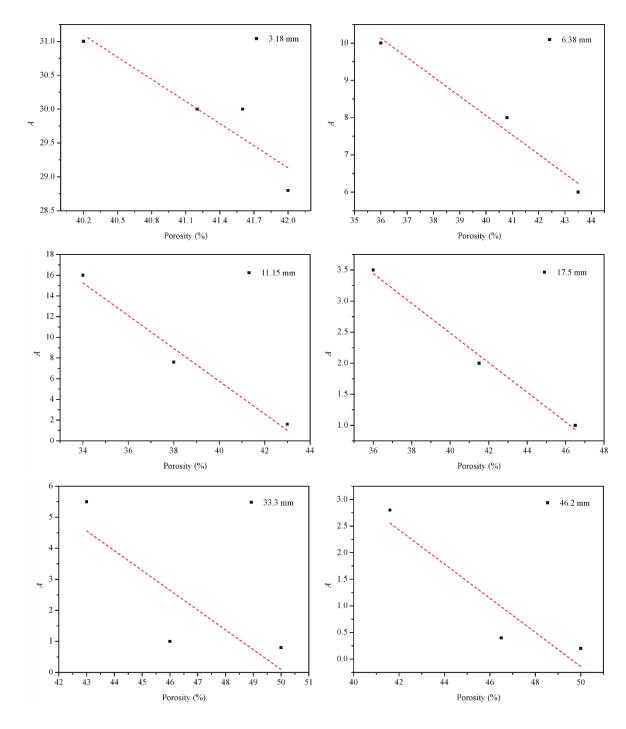


Fig. 16. The schematic diagram of cube and hexahedron arrangement with the same particle
size.

523 Table 6. Characteristic value of pore structure in different arrangement with the same particle524 size.

Arrangement mode	Side length	Porosity (%)	Specific surface area
Cube	2 <i>d</i>	47.60	3.142
Hexahedron	1.577 <i>d</i>	43.30	3.402

However, the structure of natural porous media is much more complex and heterogeneous than what has been shown in Figure 16, so it is difficult to quantitatively describe the effect of sorting degree and arrangement on seepage law.





529

Fig. 17. Variation of A with n of six gravels with different particle sizes.

In view of this, we can use a macro parameter porosity (*n*) to reveal the effect of sorting degree and arrangement on seepage coefficient. In order to verify the correctness of the above analysis results, we selected the seepage experiment results of <u>Niranjan (1973)</u> for further validation. <u>Niranjan (1973)</u> chose gravel of the same size but different porosity and carried out seepage experiments. We selected the experimental results of six different particle sizes with 3.18 mm, 6.38 mm, 11.15 mm, 17.5 mm, 33.3 mm and 46.2 mm from Niranjan (1973), and drew the relationship between coefficient *A* and *B* and porosity respectively, as shown in Fig. 17 and Fig. 18. We can see that the coefficients *A* and *B* of the six groups of experimental data of Niranjan (1973) decrease with the increase of porosity, which is consistent with our theoretical analysis of this investigation.

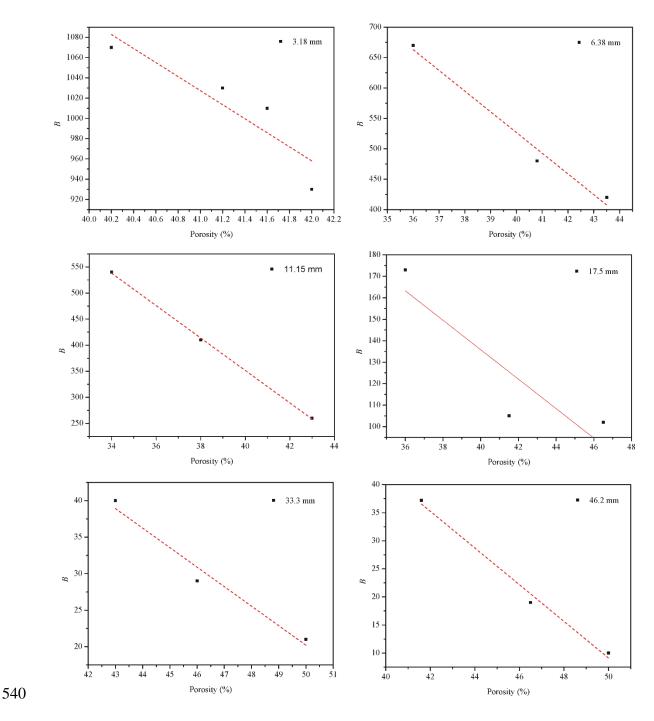




Fig. 18. Variation of *B* with *n* of six gravels with different particle sizes.

542 **4. Summary and conclusions**

This study presents experimental results of Forchheimer flow in four different permeable stones with different mesh sizes, including 24 mesh size (0.71 mm), 46 mesh size (0.36 mm), 60 mesh size (0.25 mm), 80 mesh size (0.18 mm). The effects of mean pore size and pore size distribution on the transition of flow regimes (from pre-Darcian to post-Darcian) are discussed. In addition, the mercury injection experiment is proposed to investigate the pore distribution of the permeable stones. In addition, the Forchheimer coefficients are specifically discussed. The main conclusions can be summarized as follows:

1) The relationships between specific discharge (q) and the "pseudo" hydraulic conductivity (*K*) (which is computed as a ratio of *q* and hydraulic gradient, *J*) of permeable stones show that deviation from Darcian flow regime is clearly visible. In addition, the critical specific discharge corresponding to the transition of flow regimes (from pre-Darcian to post-Darcian) increases with the increase of mean particle size.

2) When the specific discharge is small, only a small fraction of the water flowing through the pores. The rest of the water adheres to the surface of the solid particles (immobile), partially blocking the flow pathways. As the specific discharge increases, more water becomes mobile and participates in flow. Hence, the "pseudo" hydraulic conductivity increases with the increase of specific discharge. When the specific discharge increases to the critical specific discharge (q_c), the "pseudo" hydraulic conductivity is maximized, and then it begins to decrease as the specific discharge continues to increase.

3) The mercury injection experiment results show that the mercury injection curve can be divided into three segments. The beginning and end segments of the mercury injection curve of the four permeable stones with different particle sizes are very gentle, while the main (or intermediate) mercury injection curve is steep, indicating that the pore size distribution falls within a narrow range, and the proportions of large pores and small pores are relatively small.

33

567	4) The port	osity decreases as the mean particle size of permeable stone increases while the			
568	mean pore diameter increases. And the porosity can reflect the influence of particle diameter,				
569	sorting degr	ree and arrangement mode of porous medium on seepage parameters. The larger			
570	porosity lead	ds to the smaller coefficients A and B under the condition of the same particle size.			
571	5) The coeff	ficient A is linearly related to $1/d^2$ and the relationship between coefficient A and			
572	$1/d^2$ is given	n as $A = 0.0025(1/d^2) + 0.003$. The coefficient <i>B</i> is not linearly related to $1/d$,			
573	instead it is	s quadratic related to $1/d$ as $B = 1.14\text{E}-06(1/d)^2 - 1.26\text{E}-06(1/d)$. The particle			
574	shape and a	arrangement of permeable stone have imposed great influences on the seepage			
575	parameters.				
576	Notation				
577	q	The specific discharge, m/d.			
578	Κ	The Hydraulic conductivity, m/d.			
579	J	The dimensionless parameter defined as hydraulic gradient.			
580	A	The Forchheimer equation coefficient (viscous force item), sm ⁻¹ .			
581	В	The Forchheimer equation coefficient (Inertia force item), s ² m ⁻² .			
582	Pc	The capillary force, <i>Pa</i> .			
583	P_{50}	The corresponding pressure value when the saturation reaches 50%, MPa.			
584	$P_{\rm A}, P_{\rm B}, P_{\rm C}$	The pressure corresponding to different stages on mercury injection curve, MPa.			
585	σ	The solid-liquid interfacial tension.			
586	θ	The wet angle between the liquid and the solid surface.			
587	r	The radius of curvature in capillary, mm.			
588	d	The particle size, mm.			

589	d_{50}	The mean particle sizes (50% by weight), mm.
590	R_m	The mean pore diameter, μm .
591	R 50	The pore diameter corresponding to the median pressure P_{50} , μm .
592	Н	The height of the peak of the mercury injection curve.
593	X_{c}	The abscissa corresponding to the peak of the curve (the pore size).
594	W	The standard variance.
595	n	The porosity.
596	J_n	The viscous force-related component.

597 J_r The inertia force-related component.

598 Authors contributions

599 Zhongxia Li: Experiment, Writing original draft. Junwei Wan: Methodology,
600 Conceptualization. Tao Xiong: Data curation, Investigation, Experiment. Hongbin Zhan:

- 601 Methodology, Writing, Review & Editing. Linqing He: Experiment, Methodology. Kun
- 602 Huang: Funding acquisition, Investigation

603 **Competing interests**

604 The authors declare that they have no conflict of interest.

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