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2 **Permeability of Uniformly Graded 3D Printed Granular Media**  
3

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8

9 **Key Points:**

- 10 • 3D printed mono-sized yet different-shaped particles of scaling morphology features  
11 from real grains are used for permeability test.
- 12 • Via more accurate area than that of the volume-equivalent sphere method, Kozeny-  
13 Carman shape factor is found insensitive to grains shapes.
- 14 • The Kozeny-Carman equation is modified using two compressed shape factors, i.e.,  
15 fractal dimension and relative roughness.  
16

## 17 **Abstract**

18 The present work explores water permeability of uniformly graded irregular grains using 3D  
19 printing with controlled shapes and fractal morphological features at low Reynold's number for  
20 viscous flow. From large amount of real 3D granular morphological data, a scaling law, in terms  
21 of fractal dimension, is found to be followed. With this universal law, sand grains with controlled  
22 fractal morphological features are generated using Spherical Harmonics, and then created using  
23 3D printing technique for water permeability tests. A modified Kozeny-Carman equation is  
24 proposed through more accurate determination of specific area, as a function of relative  
25 roughness and fractal dimension, than approximation using the volume-equivalent sphere. By  
26 isolating the contributions from specific area, the shape coefficient is found to be insensitive to  
27 particle morphology. Through benchmarking the model prediction against experiments from  
28 both this work and past literature, we demonstrate the validity and wide applicability of the  
29 modified Kozeny-Carman equation.

## 30 **Plain Language Summary**

31 The shape of grains can influence the way how water transports inside a granular material. This  
32 study uses 3D printing technique to well control grain shapes. The grain morphology is captured  
33 through combinations of relative roughness and fractal dimension, relevant to a wide range of  
34 geomaterials. We revisit the classical permeability equation with the prior knowledge of the  
35 grain shape. It is found that the model coefficients used in the permeability equation, usually  
36 obtained through fitting against experiment measurement, do indeed contain key intrinsic  
37 morphological information.

38

## 39 **1. Introduction**

40 Estimation of permeability of porous media is of vital importance in many geophysics-  
41 related scientific fields, such as hydrogeology, geotechnical engineering, earth science, and  
42 petroleum engineering. As a main category of porous media (Higdon and Ford, 1996), granular  
43 materials received more attention owing to their significant prevalence in various engineering  
44 applications. Granular porous media are composed of solid matrix skeleton formed by bounded  
45 or contacting grains, and pores with narrow constraints. Natural and artificial examples are  
46 widely encountered in weakly cementitious rocks in sandstone (Sun et al., 2011), sands in  
47 geotechnical structures (Chapuis, 2004), fault gouge of seismic events (Wang et al., 2019), and  
48 shale soils around multilayered reservoirs (Zheng et al., 2018). For these media, permeability is a  
49 key parameter for describing their transport and hydro-mechanical responses. Seelheim (1880)  
50 stated that in porous media flow permeability correlates to the square value of its characteristic  
51 pore diameter. Since then, many models for predicting permeability coefficients have been  
52 proposed based on empirical relations (Hazen, 1892; Shepherd, 1989), Poiseuille capillary tubes  
53 (Carman, 1956; Mortensen et al., 2005), statistical regression (Wang et al., 2017; Feng et al.,  
54 2019) and effective hydraulic radius (Carman, 1956; Costa, 2006). Due to the complexity of pore  
55 network, it is challenging to rigorously formulate a satisfactory theoretical relation between  
56 viscous flow resistance and intrinsic geometry properties of granular porous media. As a result,  
57 most of such theoretical models start with some measurable intrinsic first-principle geometrical

58 factors, including sieve-determined grading, void ratio, and porosity (Hazen, 1892; Johnson et al.,  
59 1986; Chapuis and Aubertin, 2003; Ren and Santamarina, 2018).

60 Among these, one of the widely adopted models is the Kozeny-Carman equation (e.g.,  
61 Carrier, 2002):

$$62 \quad k = \frac{\gamma}{\mu} \cdot \frac{1}{C_{K-C}} \cdot \frac{1}{S_0^2} \cdot \frac{\varepsilon^3}{(1-\varepsilon)^2}, \quad (1)$$

63 where  $\gamma$  and  $\mu$  are unit weight and viscosity of permeant, respectively,  $C_{K-C}$  the Kozeny-Carman  
64 shape coefficient,  $S_0$  specific surface area (i.e., surface area per unit volume of particles),  $\varepsilon$   
65 porosity,  $k$  (L/T) the coefficient of permeability, or hydraulic conductivity, depending on both  
66 intrinsic properties of porous media and fluid, and  $K$  (L<sup>2</sup>) the permeability only correlated with  
67 porous structures. For water at 20 °C,  $\frac{\gamma}{\mu}$  equals  $9.93 \times 10^4$  1/cm s. Intuitively, contacts between  
68 grains may result in decreasing wet area, through which water flows. However, contact area is  
69 negligible when compared with the total surface area, especially for natural rough particles (Wei  
70 et al., 2020). Accompanied with Kozeny-Carman equation (Carman, 1956), the concept of  
71 tortuosity, defined as the length ratio of effective flow path to porous sample in direction of flow,  
72 is also proposed to indicate dimensionless length of fluid element flowing through pore space  
73 (David, 1993; Ghanbarian et al., 2013). Grains with the same shapes and porosity can form  
74 rather random packings, and thus different tortuosity. Such randomness may hinder the  
75 generality of tortuosity in deterministic models to predict permeability coefficients. Although  
76 many assumptions have been implemented in the original derivation of Kozeny-Carman equation,  
77 its agreement with experimental results has been widely observed (Philipse and  
78 Pathmamanoharan, 1993; Chikhi et al., 2014). As said by Wolfgang Pauli, “God made the bulk;  
79 the surface was invented by the devil”. The accurate measurement of surface area in porous  
80 media can be difficult, since many solid surfaces can exhibit multiscale features. (Barclay and  
81 Buckingham, 2009; Mollon and Zhao, 2012). With the emergent of various techniques to  
82 estimate  $S_0$ , as discussed in the review paper by Santamarina et al. (2000), a wide range of  $C_{K-C}$   
83 have been observed for grains of different morphology features. The value of  $C_{K-C}$  is found to be  
84 around 5 for monodispersed spheres (Carman, 1937). For peat beds and mixture of fibrous and  
85 granular beds, the corresponding  $C_{K-C}$  are 3.4 and 12.81, respectively (Mathvan and  
86 Viraraghavan, 1992; Li and Gu, 2005). To exclude the effects of particle grading on estimating  
87  $C_{K-C}$ , investigations are carried out using mono-sized aspherical aggregates of identical shapes,  
88 such as symmetric spiky combinations of spheres and ellipsoids (Malinouskaya et al., 2009;  
89 Thies-Weesie et al., 1995), while in reality shapes of each sand are not identical.

90 Particle shape can be described at three distinctive yet correlated length scales, namely,  
91 aspect ratio for particle size, roundness for local corner, and the finest roughness (Barrett, 1980).  
92 Dimensionless shape parameters have also been defined across these three scales. Castillo et al.  
93 (2015) experimentally estimated  $C_{K-C}$  of two types of super-ellipsoid-like cubes, i.e., solid  
94 hematite cubes and hollow microporous silica cubes, of which sizes and shapes are similar, but  
95 roughness features are different. They found that  $C_{K-C}$  of rough microporous cubes was about 12%  
96 higher, revealing the notable effects of microscopic roughness on it. With the development of  
97 optical equipment for high-resolution 3D surficial data, finding a shape index existing in more-  
98 or full-length morphology is possible. Fractal dimension, accompanied by relative roughness, is

99 a candidate, which may unify global surface morphology across multiple length scales (Barclay  
100 and Buckingham, 2009; Renard et al., 2013).

101 In this work, the effect of particle shapes on permeability of porous media is  
102 experimentally investigated via hydraulic conductivity tests, as done in classical soil mechanics  
103 (Craig, 2004). Realistic particles with controlled fractal dimension are generated using Spherical  
104 harmonics (Wei et al., 2020), which are then printed using 3D printers. A modified Kozeny-  
105 Carman equation is proposed by incorporating the fractal dimension and relative roughness, two  
106 key morphological parameters for a wide range of geomaterials. Through comparing the model  
107 with experimental results and data from literature, we then assess the ability of the proposed  
108 model to capture the influence of particle shapes on permeability of granular materials.

109

## 110 2. Spherical harmonic reconstruction

### 111 2.1. Reconstruction of natural grains

112 For spherical surfaces, through 3D Fourier Transformation, any function set on the  
113 surface can be represented as a sum of Spherical Harmonic (SH) function,  $Y_n^m(\theta, \varphi)$ , for its  
114 orthogonality. SH function has been dedicated to many scientific applications, such as  
115 representing orbital configurations (Flügge, 2012), computing physical fields (Turcotte, 1987),  
116 and modelling 3D images (Garboczi, 2002). Here, we implement its definition in quantum  
117 mechanics to reconstruct star-like surfaces of natural grains via approximating its cumulative  
118 radial distance,

119  $r_I(x_I(\theta, \varphi), y_I(\theta, \varphi), z_I(\theta, \varphi)) = r_I(\theta, \varphi) = \sqrt{(x_I - x_0)^2 + (y_I - y_0)^2 + (z_I - z_0)^2}$ , between  
120 surface points and its centroid,  $(x_0(\theta, \varphi), y_0(\theta, \varphi), z_0(\theta, \varphi))$ , as a function of latitudinal ( $\theta \in$   
121  $[0, \pi]$ ) and longitudinal ( $\varphi \in [0, 2\pi]$ ) coordinates in polar coordinate system:

$$122 r_I(\theta, \varphi) = \sum_{n=0}^{\infty} \sum_{m=-n}^n c_n^m Y_n^m(\theta, \varphi), \quad (2)$$

123 where  $I$  denotes the  $I$ -th point on particle surface and  $c_n^m$  are the SH coefficients of degree  $n$  and  
124 order  $m$ . The process to calculate complex  $c_n^m$  is in S1. Fig. 1 (a) shows nine types of real  
125 granular materials of wide-scoped sizes, and the scaling exponential relations in Fig. 1 (b) read

$$126 D_n \propto n^\beta, (n = 2, 3, 4, 5 \dots), \quad (3)$$

127 where  $D_n$  is the SH descriptor defined as normalised  $L2$  norm ( $D_n = \frac{\sqrt{\sum_{m=-n}^n \|c_n^m\|^2}}{c_0}$ ),  $\beta$  is the slope  
128 of the regression plot of  $\log(D_n)$  versus  $\log(n)$ . Consequently, following Russ (1994) and  
129 Quevedo et al. (2008),  $D_n$  can be expressed in terms of SH fractal dimension ( $D_f$ ):

$$130 D_n = D_2 \cdot \left(\frac{n}{2}\right)^{2D_f - 6}. \quad (4)$$

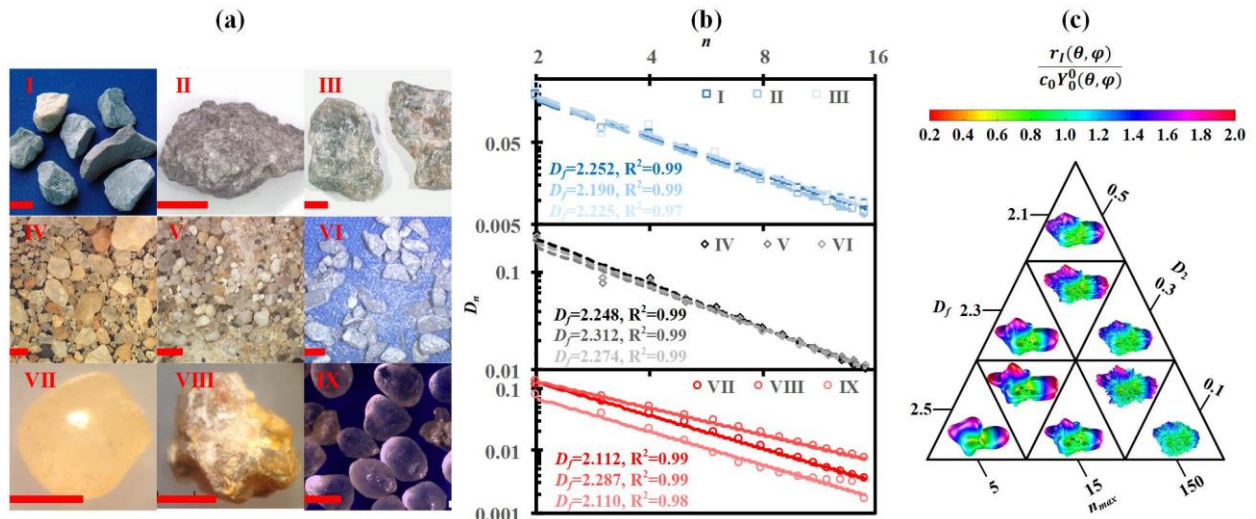
131 According to the Parseval formula, the mean square distance ( $M_{SD}$ ) between two SH surfaces  
132 with SH coefficient  $c_{1,n}^m$  and  $c_{2,n}^m$  can be directly computed (Gerig, 2001):

133  $M_{SD} = \frac{1}{4\pi} \sum_{n=0}^{\infty} \sum_{m=-n}^n \|c_{1,n}^m - c_{2,n}^m\|^2$ . Then, the relative roughness ( $R_r$ ), quantifying how the  
134 irregular particle surface is globally different from its  $c_0$ -determined sphere, can be defined  
135 based on  $\sqrt{M_{SD}}$ :

$$R_r = \frac{\sqrt{\frac{1}{4\pi} \sum_{n=2}^{n_{\max}} \sum_{m=-n}^n \|c_n^m\|^2}}{c_0^0 \cdot Y_0^0(\theta, \varphi)} = \sqrt{\sum_{n=2}^{n_{\max}} \left( D_2 \cdot \left( \frac{n}{2} \right)^{2D_f - 6} \right)^2}. \quad (5)$$

137 Details of derivation and determination of  $D_f$  and  $R_r$  are provided in S2. S3 is referred to  
 138 for the method of generating randomly shaped particles of given  $D_2$  and  $D_f$  via stochastic  $c_n^m$ . It  
 139 has been confirmed that virtual and real particle shapes can have nearly identical shape  
 140 parameters (e.g., the difference between their mean parameters are all within 2 %), as long as  
 141 their corresponding  $D_2$  and  $D_f$  are the same (Wei et al., 2018).

142



143 **Figure 1.** The framework from experimentally scanned grains to virtually generated particle  
 144 shapes. (a) Nine types of real aggregates with scale bar being 1 cm for I to III and 1mm for IV to  
 145 IX. According to American Society for Testing and Materials (ASTM), the corresponding names  
 146 of aggregates from I to VI are MA106A-1, MA107-6, MA114F-3, MA111-7, MA99BC-5 and  
 147 MA106B-4. Images are obtained from an open source software, Virtual Cement and Concrete  
 148 Testing Laboratory (VCCTL, Bullard, 2014); VII and VIII, Leighton Buzzard sand and highly  
 149 decomposed granite are from Wei et al. (2018); IX, Ottawa sand, is from Erdoğan et al. (2017).  
 150 (b) Relations between average  $D_n$  and  $n$  in log-log scales for grains in (a). (c) Virtual  
 151 representation of generated particle shapes using various maximum SH degree,  $n_{\max}$ ,  $D_f$  and  $D_2$ .  
 152 The colour bar represents the ratio of normalized radial distance to radius of its  $c_0$ -determined  
 153 sphere.

154

## 155 2.2. Approximation of volume and surface area

156 Since the surface can be implicitly approximated by SH expansion in Eq. (1), grain  
 157 volume ( $V$ ) and surface area ( $S$ ), directly relevant to  $S_0$  in Kozeny-Carman equation, can be  
 158 computed and denoted by  $c_n^m$ . For SH-generated surface of maximum SH degree,  $n_{\max}$ , one  
 159 particle has  $(n_{\max} + 1)^2$  complex numbers. We set  $n_{\max}$  to 15, which is sufficient in depicting  
 160 morphology features finer than roundness-length scale of  $n_{\max} = 8$  (Zhao et al., 2018). Notably,  
 161 as shown in Figure 1 (c), the applied  $n_{\max}$  scales for depicting rougher grain morphology. Via  
 162

163 comparing the surface area between SH-approximated and CT (computer tomography)-based  
 164 grain shapes, Zhou et al. (2017) found the approximate  $n_{\max}$  for rough HDG particles are higher  
 165 than smooth LBS particles. Computational wise, it is expected there is always space to improve,  
 166 so  $n_{\max}$  can be higher. However, the length-scale of input morphology features is limited by 3D  
 167 printing resolution. Further reasons for selecting  $n_{\max} = 15$  as the cut-off length to calculate  $S_0$   
 168 can be found in Sections 3 and 4.

169 Considering the volume element in polar coordinate system,  $dV = r \sin \theta d\theta \cdot r d\varphi \cdot dr$ ,  $V$   
 170 can be rigidly determined,

$$171 \quad V = \int_0^{r(\theta, \varphi)} \int_0^\pi \int_0^{2\pi} r(\theta, \varphi)^2 \sin \theta d\varphi d\theta dr. \quad (6)$$

172 By inserting Eq. (2) into Eq. (6),

$$173 \quad V = \underbrace{\frac{c_0^3}{6\sqrt{\pi}}}_{V_{n=0}} + \underbrace{\frac{3c_0}{6\sqrt{\pi}} \cdot \sum_{n=2}^{n_{\max}} \left( D_2 \cdot \left( \frac{n}{2} \right)^{2D_f - 6} \right)^2}_{\Delta V} = \frac{c_0^3}{6\sqrt{\pi}} (1 + 3R_r^2), \quad (7)$$

174 where  $V_{n=0}$  is the volume of its  $c_0$ -determined sphere with radius,  $c_0^0 \cdot Y_0^0(\theta, \varphi) = \frac{c_0^0}{2\sqrt{\pi}}$ ,  $\Delta V$  is the  
 175 changed volume of  $V$ . To check the integration accuracy of Eq. (6), 2,000 virtual shapes are  
 176 generated for various sets of  $D_f$  and  $R_r$ , and the mean  $\Delta V(\mu_{\Delta V})$  can be calculated. It is found that  
 177 the ratios of standard deviation to its corresponding  $\mu_{\Delta V}$  are less than 0.05. From figs. 2 (a) and  
 178 (b),  $V$  does depend on  $D_f$ , while the dependence can be entirely caught by  $R_r$ , indicating the  
 179 reasonableness of Eq. (7).

180 A surface element on curved surface is based on its local differential properties in  
 181 Cartesian coordinate system:

$$182 \quad dS = |\vec{X}_\theta \times \vec{X}_\varphi| d\theta d\varphi, \quad (8)$$

183 where  $\vec{X} = (x, y, z)$  is the surface normal vector, and subscripts denote partial differential items.  
 184 The unit surface normal vector is  $\frac{\vec{X}_\theta \times \vec{X}_\varphi}{|\vec{X}_\theta \times \vec{X}_\varphi|}$ , the components of which are expanded in S4.  
 185 Accordingly, the analytical solution of SH-generated surface area is

$$186 \quad S = \int_0^\pi \int_0^{2\pi} r \cdot \sqrt{(r_\varphi^2 + r_\theta^2 (\sin \theta)^2 + r^2 (\sin \theta)^2)} d\varphi d\theta, \quad (9)$$

187 where

$$188 \quad r_\theta = -\sum_{n=0}^{\infty} \sum_{m=-n}^n \sqrt{\frac{(2n+1)(n-m)!}{4\pi(n+m)!}} \frac{c_n^m}{\sin \theta} [(n+1) \cos \theta P_n^m + (m-n-1)P_{n+1}^m] e^{im\varphi}, \quad (10)$$

189 and

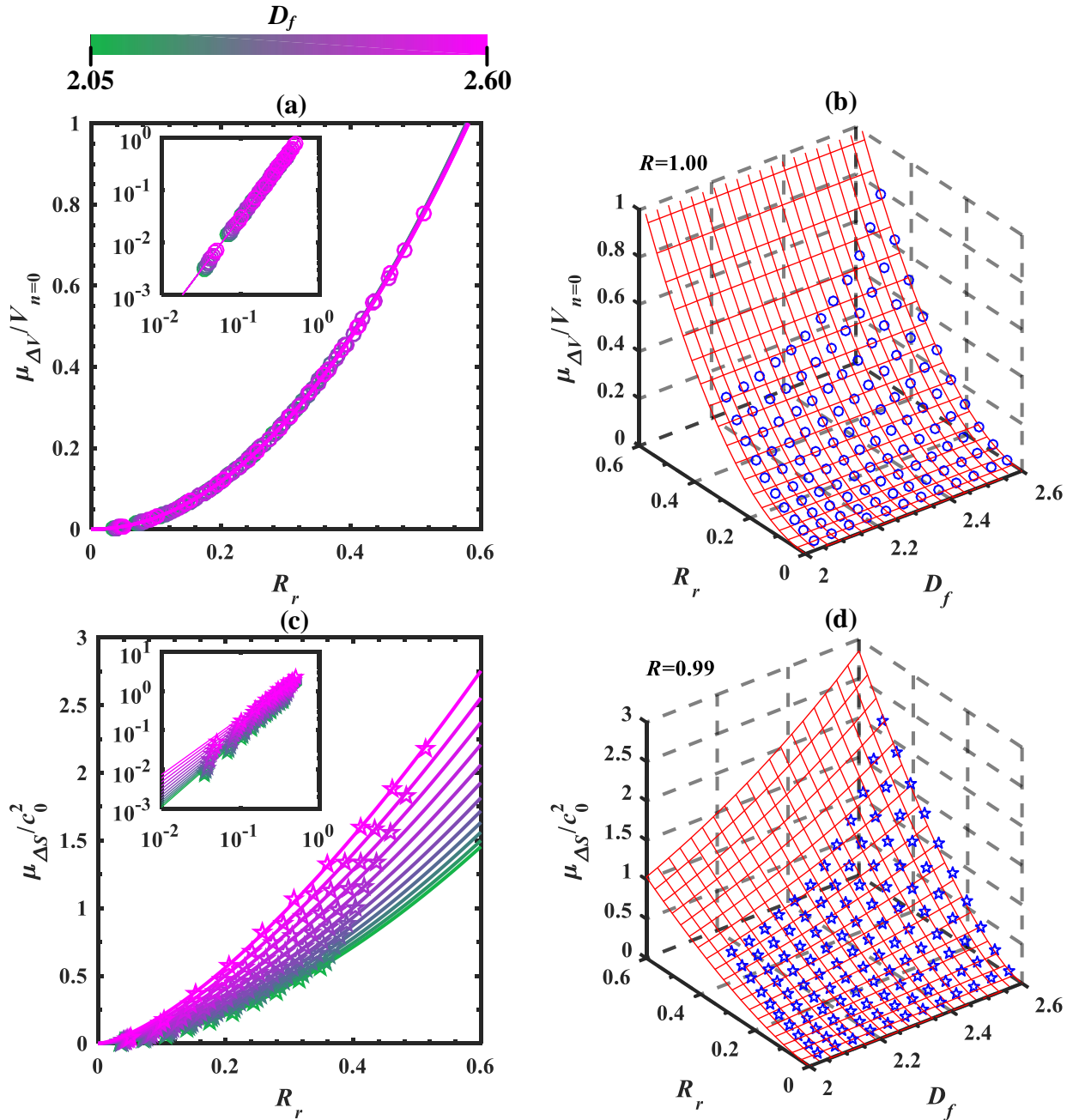
$$190 \quad r_\varphi = \sum_{n=0}^{\infty} \sum_{m=-n}^n m \cdot i \cdot c_n^m Y_n^m(\theta, \varphi). \quad (11)$$

191 Although in Eq. (9) a closed-form expression of  $S$  is derived, the representation is too  
 192 complicated to directly relate to  $D_f$  and  $R_r$ . Similar to the treatment with  $V$ ,  $S$  is split into two  
 193 parts:  $S = S_{n=0} + \Delta S$ . From Fig. 2 (c) and Fig. S1, power laws between the mean  $\Delta S$  ( $\mu_{\Delta S}$ ),  $D_f$   
 194 and  $R_r$  can be clearly seen. As a simplification, regression analysis is used to define  $S$  as a  
 195 combined function of the two,

$$196 \quad S = \underbrace{c_0^2}_{S_{n=0}} + \underbrace{c_0^2 \cdot \frac{\pi}{20} \cdot R_r^{\frac{\pi}{2}} \cdot D_f^{3.874}}_{\Delta S} = c_0^2 \left( 1 + \frac{\pi}{20} \cdot R_r^{\frac{\pi}{2}} \cdot D_f^{3.874} \right). \quad (12)$$

197 Fig. 2 (d) indicates the efficiency of Eq. (12) in approximating  $S$ . Notably, the ranges of  $R_r$  or  $D_2$   
 198 and  $D_f$  ( $D_2 \in [0, 0.3]$ ,  $D_f \in [2, 2.6]$ ), in which the regression is conducted, are much larger than  
 199 those of real sands, as in Fig. 1 (b). Considering the high value of R-square, it is reasonable to  
 200 directly apply Eq. (12) to approximate surface area of SH-generated surfaces in subsequent  
 201 studies.

202



203



204 **Figure 2.** Influences of  $D_f$  and  $R_r$  on mean varied  $V$  and  $S$ , of which every data point is from  
 205 2,000 virtual particles of  $n_{\max} = 15$ . The morphology features smaller than cut-off length does not  
 206 contribute to  $S_0$ .  $\circ$  and  $\star$  denote  $\mu_{\Delta S}$  and  $\mu_{\Delta V}$  from closed form solutions from Eq. (6) or Eq. (9).  
 207 The solid line and mesh in (a) and (b) denote solutions from Eq. (7), while the solid line and  
 208 mesh in (c) and (d) are from regression analysis. For every data point of  $\mu_{\Delta S}$  and  $\mu_{\Delta V}$ , the ratio of  
 209 its standard deviation to itself is less than 0.05. The efficiency of the developed regression  
 210 conducted to approximate  $\mu_{\Delta S}$  depends on the number of training data. It is found that when the  
 211 number of particle shapes for every point data in (d) is larger than 2000, the resulted parameters  
 212 in Eq. (12) vary less than 1 %.

213

### 214 3. 3D printed grains and water permeability experiments

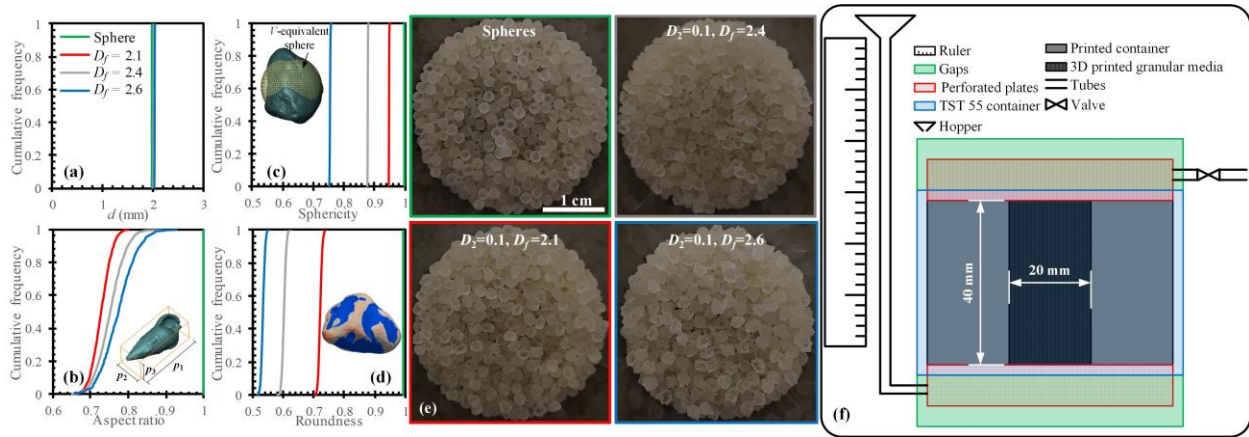
215 In recent years, for the capability of producing particles with controlled morphology and  
 216 material properties, 3D printing (Jiang et al., 2020) are becoming popular in studies of granular  
 217 mechanics (Miskin and Jaeger, 2013; Ju et al., 2018; Gupta et al., 2019) and hydraulic  
 218 conductivity (Suzuki et al., 2017; Fang et al., 2018; Adamidis et al., 2019). However, many  
 219 studies just consider regular aspherical shapes for idealization (Athanasiadis et al., 2014;  
 220 Murphy et al., 2019). Meanwhile, for irregular shapes most 3D printed particles are produced as  
 221 one-to-one of CT data for estimating whether they can capture real granular behaviour. As  
 222 pointed by the pioneering work on 3D printable geomaterials (Hanaor et al., 2016), except for  
 223 printing resolution and materials, progress can be dedicated to geometrical model of printed  
 224 grains. The following advantages can be obtained: i) input morphological and statistical  
 225 parameters are extracted from real ones; ii) as many as desired realistic particle morphology  
 226 features are depicted, such as coexistence of smoothed faces, angularity and branching; iii) ready  
 227 to study effects induced by varying morphology. Looking back to Fig. 1, all of the mentioned  
 228 advantages can be achieved via our proposed SH framework.

229 In this work, a poly-jet layer-printing 3D printer (Objet Eden 250) with horizontal and  
 230 vertical resolutions of 4  $\mu\text{m}$  and 32  $\mu\text{m}$  was used. For more information on the printer and  
 231 printing materials, please refer to the relevant study (Hanaor et al., 2016). Each batch containing  
 232 2,000 particles, placed by 40 rows and 50 columns in one layer, is printed. As suggested by  
 233 Adamidis et al. (2020), with the help of 3D printing (the resolutions are 16  $\mu\text{m}$  and 40  $\mu\text{m}$  in  
 234 horizontal and vertical resolutions), shape parameters of printed grains of size equal around 2mm  
 235 are within 5 % difference from input geometries, proved by X-Ray CT images. With SH  
 236 coefficients from section 2.1, we import polar coordinates of icosahedron-based geodesic  
 237 structure with 1,280 triangular faces into Eq. (2) to generate STereolithography (STL) input files  
 238 suitable for 3D printing. The reason why surfaces composed of 1,280 faces are selected is that  
 239 the triangle edge length should be larger than the printing resolution for printing all given  
 240 geometry features. Edges connecting adjacent vertices of such surficial meshes have a spherical  
 241 angle of about 0.14. The depicted grain morphology is influenced by both the number of vertices  
 242 on the surface and  $n_{\max}$ . Considering the relation between a proper angle resolution ( $\Delta\theta$ ) and  
 243 any given  $n$  (Jekeli, 1996; Jekeli et al., 2007),  $\Delta\theta = \frac{\pi}{n}$ , and the vertical printing resolution, the  
 244 finest morphology features, which can be successfully printed, are determined by  $n_{\max}$  up to  
 245 about 30, since  $r_l(\theta, \varphi) \approx 1 \text{ mm}$ . This being said, there are two main reasons why  $n_{\max} = 15$  is  
 246 selected: i) when removing supporting wax after printing, NaOH solution may corrode finer

247 grain surface features; ii) for  $D_f < 2.75$ , when  $n_{\max} = 15$ , in S6 it is proved that  $R_r$  depending  $S_0$   
 248 has converged and is less than  $\frac{2^{6-2D_f} \cdot D_2}{\sqrt{11-4D_f}}$ . Furthermore, we align the longest particle dimension  
 249 with vertical direction to optimally exploit the printing resolution.

250 Fig. 3 (a)-(d) illustrates the cumulative distributions of input shape indices for four  
 251 groups of particles with the same  $c_0 = 3.5$  mm. The non-uniformly shaped particles have  
 252  $D_2 = 0.1$ , close to those of fine aggregates in Fig.1 (a) and (b). It can be seen that the printed  
 253 particles have uniform gradings (Fig. 3(a)), and their sphericity is also uniform within a group,  
 254 consistent with Eqs. (5), (7), and (12). The variation in aspect ratio indicates the efficiency of the  
 255 proposed SH framework in generating macroscopically different particle shapes yet with  
 256 statistically similar finer morphological features, which is further proved in Fig. 3 (d) by the  
 257 median-sized feature, roundness. After finishing printing, the printed grains with supporting wax  
 258 were kept in a bath with aqueous solution containing 2 % NaOH for 30 minutes. Then, the grains  
 259 were manually rubbed in the basin to remove remaining wax, and finally rinsed with pure water.  
 260 At last, wetted grains were dried overnight at constant room temperature of 20 °C, as in Fig. 3 (e).

261



262

263 **Figure 3.** (a)-(d) Cumulative distributions of classical shape indices of printed grains of  $c_0=3.5$   
 264 mm ( $d \approx 2$  mm) and  $D_2 = 0.1$ . Each group contains about 10,000 particles. (a) Equivalent-sphere  
 265 diameter. (b) Aspect ratio, defined as  $A_r = E_i \times F_i$ , with mean elongation and flatness  $E_i =$   
 266  $p_2/p_1$  and  $F_i = p_3/p_2$ , respectively, where  $p_1$ ,  $p_2$  and  $p_3$  are the particle's principal dimensions  
 267 calculated by principal component analysis. (c) Sphericity  $S_p = \sqrt[3]{36\pi V^2/S}$  reflects the  
 268 deviation of surface area from its volume-equivalent sphere. Notably,  $S_p$  can be used to indicate  
 269 how much error can be made if the area of grain volume-equivalent sphere is considered to  
 270 approximate its surface area. For example, if the area of volume-equivalent sphere is applied,  
 271 about 25% error can be induced. (d) Roundness  $R_M = \sum \left( S_l \cdot \frac{k_{in}}{k_{M,l}} \right) / \sum S_l, \forall k_{M,l} \leq k_{in}$ , where  $S_l$   
 272 is the area of  $l$ -th triangle, of which the mean median curvature value of its three vertices is  $k_{M,l}$ ,  
 273 and  $k_{in}$  is the curvature value of maximum inscribed sphere of the particle. The colour in (d)  
 274 represents mean median curvature value of composed triangles. (e) Snapshots of printed grains.  
 275 (f) Schematic of the modified TST-55 permeameter for experiments.

276

277 Capabilities of one-to-one printed sands to reproduce hydraulic-related features have  
 278 been comprehensively demonstrated by Adamidis et al (2020). The standard TST-55  
 279 permeameter (GB/T 9357-2008) was used for water permeability tests via falling-head method.  
 280 Since the volume of original container of TST-55 permeameter is too large to be completely  
 281 filled with our printed grains, a hollow cylinder was printed as a filler to reduce the effective  
 282 volume of the original container (Fig. 3(f)). The ratio of average grain diameter to container  
 283 width is about 0.1, which is sufficiently small to obtain representative measurements of hydraulic  
 284 conductivity (Garcia et al., 2009). For permeability test, grains were poured into the container,  
 285 and the excessive grains were carefully levelled off. To vary the porosity, different intensity of  
 286 tapping and compression were applied during pouring. As pointed out by Chapuis (2012), errors  
 287 in laboratory tests of the hydraulic conductivity mainly fall into two categories: i) the occurrence  
 288 of preferential leakage between the porous specimen and the rigid permeameter wall; ii) the full  
 289 saturation of the porous media is not achieved. To avoid the former, the inner surface of the  
 290 printed hollow cylinder is mapped by closely wounded coils with sectional circles of diameter  
 291 equal to 1 mm. For the latter, the permeameter was vibrated during first several cycles of water  
 292 injection until no observable bubbles can be seen. By slowly seeping the fluid from the base to  
 293 the top, the gravity is also in favour of removing the air. In Taylor et al. (2017), hydraulic  
 294 conductivity of natural LBS particles is conducted by seepage from the base, of which the results  
 295 are also compared with samples with de-air process. It was found the difference between the two  
 296 is within 2 %. For subsequent studies, the application of degassed water is a good choice. Via  
 297 repeating the tests for a given experiment, it is found that the difference of measured coefficients  
 298 of permeability is also within 2 %.

299 Videos at 30 fps were recorded during permeability tests. Compared with classical  
 300 falling-head test, where only one time period ( $t_{n+1} - t_n$ ) and its two water head heights ( $h_n$  and  
 301  $h_{n+1}$ ) are considered. The hydraulic conductivity is obtained as,

$$302 \quad k = \frac{a \cdot l}{A \cdot (t_{n+1} - t_n)} \cdot \ln \frac{h_n}{h_{n+1}}, \quad (13)$$

303 then we modify it as

$$304 \quad k_n = \frac{a \cdot l}{A} \cdot \frac{\ln h_n - \ln h_{n+1}}{t_{n+1} - t_n}, \quad (14)$$

305 where  $a$  and  $A$  are the areas of the tube and the porous sample cross-section, respectively, and  $n$   
 306 is the frame number of the video. By using all the data during each permeability test (typically  
 307 several hundreds of frames),  $k$  can be determined by fitting the slopes in  $(\ln h)$ - $t$  curves. Seven  
 308 representative experiments were conducted for each group of grain type at each porosity, and the  
 309 invariance to mean ratio is less than 5%, indicating good repeatability of experiments.

#### 310 4. Results and modified Kozeny-Carman equation

311 Fig. 4 (a) shows the measured absolute coefficients of water permeability,  $K$ , as a  
 312 function of porosity  $\varepsilon$  for different particle shapes. With the increase of  $D_f$ , the tortuosity of pores  
 313 in porous media of same  $\varepsilon$  becomes higher, resulting in lower  $K$ . Some studies have revised  
 314 Kozeny-Carman equation based on the specific grading to approximate pore diameter, such as in  
 315 Chapuis (2004) with  $k = 2.4622 \cdot [D_{10}^2 \cdot \frac{\varepsilon^3}{(1-\varepsilon)^2}]^{0.7825}$ , where  $D_{10}$  is the 10% finer particle size.

316 This approach is not applicable to the current work, because the gradings of our four types of  
 317 printed grains are deemed equal, as shown in Fig. 3 (a).

318 Since printed grains are reconstructed along vertices of 1280-triangle based icosahedron  
 319 geodesic structures,  $S$  and  $V$  can be directly calculated and the corresponding  $S_0$  can be readily  
 320 obtained. Fig. 4 (b) compares  $C_{K-C}$  calculated by importing measured permeability into Eq. (1). It  
 321 is surprising that the shape factor,  $C_{K-C}$ , is insensitive to particle shapes from spheres to highly  
 322 irregular shapes with  $D_f = 2.6$ . However, if  $S_0$  is approximated based on volume-equivalent  
 323 spheres, an increase in  $C_{K-C}$  is observed for irregular particles. Here, we propose that the increase  
 324 is the manifestation of the errors from calculation of  $S_0$ . The value of  $C_{K-C}$  of printed spheres is  
 325 about 5.5, within the relevant experimental values 5.0 ~ 6.4 for mono-sized spheres (Carman,  
 326 1956). This consistency in  $C_{K-C}$  of mono-sized spheres proves the adequacy of our experiments  
 327 to measure permeability.

328 Via electrokinetic analysis of complex anion adsorption, Hanaor et al. (2014) concluded  
 329 that particle surface area is unlimited. If so,  $K$  would approach to zero, which is certainly  
 330 problematic. According to Koch and Brady (1985) and Durlofsky and Brady (1987), the decay  
 331 length, called Brinkman screening length, of viscous flow velocity disturbance in fixed spherical  
 332 granular porous media roughly equals  $\sqrt{K}$ . This decay length can be regarded as the cut-off  
 333 length for effects of surface roughness on the near flow field, below which surface features will  
 334 have limited effect on the permeability (Castillo et al., 2015). Thus, only morphological features  
 335 of length beyond such length is considered into  $S_0$ . Note that the roughness of printed grains is  
 336 deemed the same and the 3D printer resolution is 32  $\mu\text{m}$ , which is in the same order of magnitude  
 337 as mean triangular edge length ( $\approx 136 \mu\text{m}$ ) of printed triangle-based grains, as well as the decay  
 338 length ( $\approx 20 \mu\text{m}$ ). We fit data points of printed spheres in Fig. 4 (b) to get  $C_{K-C}$  for the roughness  
 339 generated by the printer. Substituting Eqs. (7) and (12) into Eq. (1), the modified Kozeny-  
 340 Carman equation is

$$341 \quad K = k \cdot \frac{\mu}{\gamma} = \frac{1}{5.5} \frac{c_0^2 (1+3R_r^2)^2}{36\pi \left(1 + \frac{\pi}{20} R_r^2 \cdot D_f^{3.874}\right)^2} \frac{\varepsilon^3}{(1-\varepsilon)^2}. \quad (15)$$

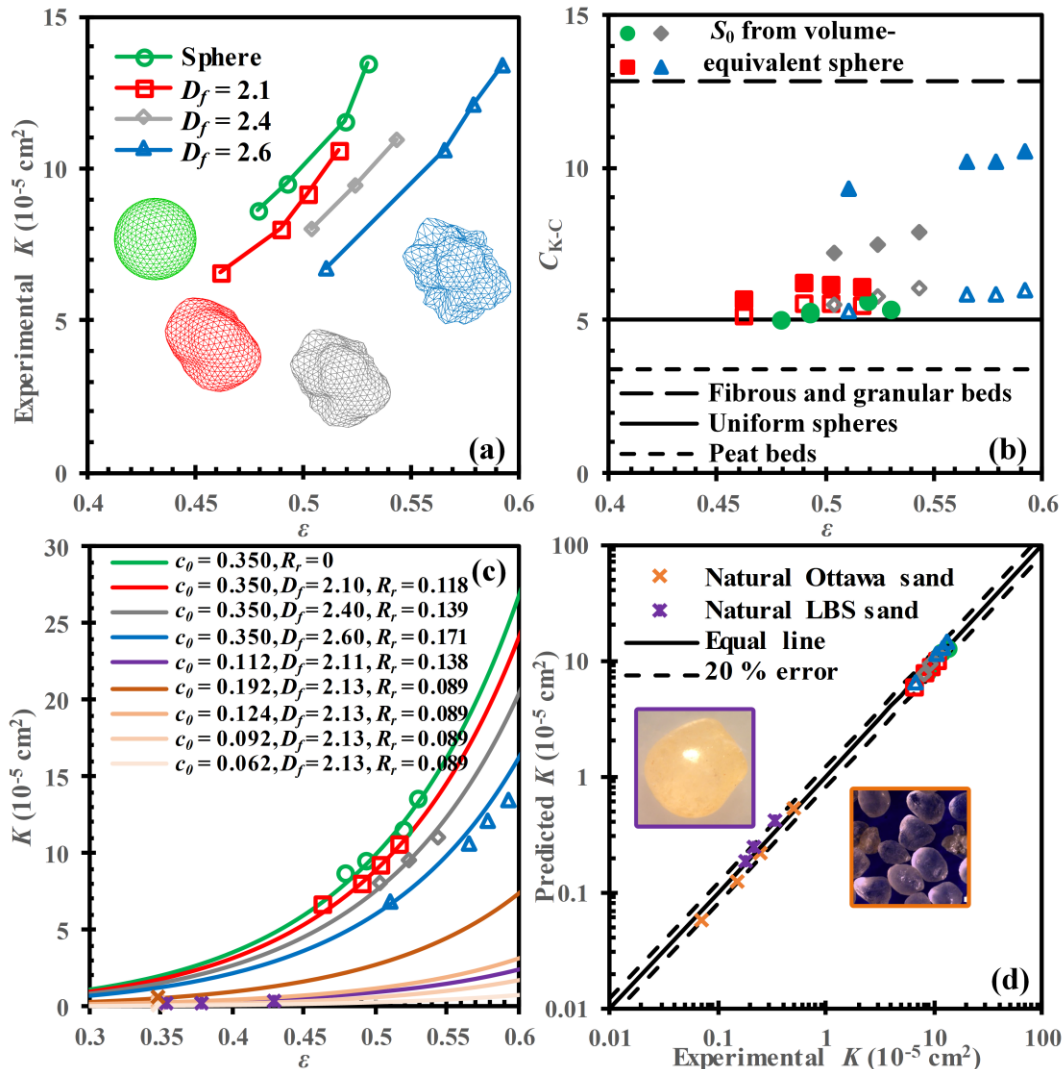
342 Notably,  $R_r$  is dependent on  $n_{\max}$ , which can be related to the decay length with  $\sqrt{K} \propto$   
 343  $\frac{\pi}{n_{\max}} r_l(\theta, \varphi) \approx \frac{\pi}{n_{\max}} c_0^0 Y_0^0(\theta, \varphi)$ ,

$$344 \quad n_{\max} \propto \frac{c_0}{2} \cdot \sqrt{\frac{\pi}{K}}. \quad (16)$$

345 Return to Eq. (5), the value of  $R_r$  converges and is less than  $\frac{2^{6-2D_f \cdot D_2}}{\sqrt{11-4D_f}}$ . In addition, from Fig. S2  
 346 when  $n_{\max}$  is up to 15, the value of  $R_r$  is very close to the convergence. Hence, we choose  $n_{\max} =$   
 347 15 to calculate  $R_r$ . After importing  $R_r$  calculated from Eq. (5),  $c_0$ , and  $D_f$  into Eq. (15),  
 348 predictions of  $K$  can be obtained, which agree well with the experimental results in Fig. 4 (c). It  
 349 proves the reasonableness of  $n_{\max} = 15$ , as a cut-off, to calculate  $S_0$  for  $K$ , at a low Reynolds  
 350 number. Note this modified equation does not contain any additional fitting parameters and only  
 351 requires porosity and morphological parameters accessible from actual grain shapes.

352 To further check applicability of Eq. (15) for natural grains, 3D surficial data (e.g., X-  
 353 Ray CT data) are needed for determination of  $c_0$ ,  $R_r$ ,  $D_f$  at  $n_{\max} = 15$ . Two types of poorly graded  
 354 natural quartz sands (LBS from Taylor et al. (2017) and Ottawa sand from Schroth et al. (1996))  
 355 are taken into consideration in Figs. 4 (c) and (d), where  $R_r$  and  $D_f$  are calculated from Fig. 1 (b),

356 sample VII and IX, respectively. Although there is no perfectly uniform grading of the two sands,  
 357 predicted values are close to experimental values. The model validated by data of natural sands  
 358 appears to be appropriate over three orders of magnitudes of water permeability coefficient. It is  
 359 possible that the proposed equation is not appropriate for well graded grains, because packing  
 360 structures or pore distributions can significantly differ from those of uniform gradings. How  
 361 grain grading distribution affects the validity of the equation deserves future studies. In this study,  
 362 only statistical morphological features ( $R_r$  and  $D_f$ ) and volume are uniform, but other shape  
 363 parameters lay in a wide range, as in Fig. 3 (b), which could also alter pore structures. This may  
 364 explain why the proposed model can hold for  $C_u$  (grading uniform coefficient) up to 1.5. To the  
 365 best knowledge of the authors, it is the first time to experimentally study permeability  
 366 coefficients of non-spherical uniform grains with focus on whether the shape coefficient in  
 367 Kozeny-Carman equation is dependent on grain shapes, when effects of roughness are isolated.  
 368 Surprisingly, the Kozeny-Carman shape coefficient is insensitive to particle shapes, contrary to  
 369 what has been widely suggested before. Although via hydraulic conductivity tests such  
 370 insensitivity is found, how much it can be generalized to other types of porous media with global  
 371 multi-scale morphology features, such as fibrous structures, needs further research.  
 372



373

374 **Figure 4.** (a) Experimental water permeability coefficients as a function of porosity. The  
 375 snapshots represent 1280-face particle surface with the same polar coordinates of icosahedron-  
 376 based geodesic surface with the same number of facets. (b) Relations between  $C_{K-C}$  and porosity  
 377 for  $S_0$  calculated from STLs (void symbols) or volume-equivalent spheres (solid symbols).  
 378 Values for fibrous and granular beds, uniform spheres, and peat beds are from Li and Gu (2005),  
 379 Carman (1937), and Mathvan and Viraraghavan (1992), respectively. (c) Relations between  
 380 porosity and water permeability coefficients with lines for the proposed equation and data points  
 381 for experimental data. The colour orange and purple denote natural Ottawa sand and LBS  
 382 particles. The unit of  $c_0$  is cm. (d) Comparisons between experimental results and predictions of  
 383 modified Kozeny-Carman equation, including two natural LBS (Taylor et al., 2017) and Ottawa  
 384 sand (Schroth et al., 1996) particles.

385

## 386 5. Conclusions

387 With the help of 3D printing, at low Reynold's number the present work explores  
 388 coefficients of permeability of uniformly graded irregular grains with controlled shapes and  
 389 fractal morphological features. The results indicate that particle shape does influence  
 390 permeability coefficients; that is, with the increase of fractal dimension only above a moderate  
 391 roughness length scale, induced high tortuosity can slow water seeping through granular porous  
 392 media of the same porosity. A modified Kozeny-Carman equation is proposed by incorporating  
 393 the specific surface area calculated based on spherical harmonics, where it is derived as a  
 394 function of particle size, relative roughness and fractal dimension. It is found that the shape  
 395 coefficient in Kozeny-Carman equation is insensitive to particle shapes, ranging from spheres to  
 396 significantly irregular shapes of high fractal dimension equal to 2.6. We also check the  
 397 applicability of the proposed model on poorly graded natural grains by comparing with data in  
 398 the literature. Good agreements are observed. The current work provides the first experimental  
 399 study on permeability of uniformly graded aspherical grains with controlled particle shapes and  
 400 fractal morphological features, and advances the understanding of their correlations. Extensive  
 401 research on how the combined effects of grain sizes and shapes affect the proposed permeability  
 402 model prediction should be performed in future studies. To better solve this outstanding issue,  
 403 appropriately quantifying stochasticity of tortuosity, which is for pore structures that fluids flow  
 404 through, may be necessary in deterministically predictive models of permeability coefficients.

405

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414

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