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Effect of grain shape on quasi-static fluid-fluid displacement in porous media

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Key	Points:
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9	•	A novel pore network model algorithm is developed to probe the effect of grain
10		shape on multiphase displacement in porous media.
11	•	Systematic simulations are conducted using the proposed algorithm across a wide
12		range of wetting conditions and particle shapes.
13	•	Through analyzing various metrics during displacement, the results highlight the
14		profound influence of particle shape on multiphase flow.

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15 Abstract

We study how grain shapes impact multiphase flow in porous media in the quasi-static 16 regime using an extended pore-network model. The algorithm allows the explicit deter-17 mination of different types of pore-scale instabilities and tracks the interface motion dur-18 ing fluid-fluid displacement process. It also includes the volume capacitance model, such 19 that both the evolution of capillary pressure signal and sizes of Haines jumps can be cap-20 tured. Further, it considers the pinning of menisci at sharp edges of grains, through which 21 the distribution of effective contact angles can be obtained. Simulations are carried out 22 across a wide range of wetting conditions for different particle shapes. Our results show 23 that the effective contact angle distribution during displacement widens as the grain be-24 comes more angular, which consequently modifies the macroscopic fluid invasion mor-25 phology. By analyzing various characteristic metrics during displacement, including cap-26 illary pressure signal, Haines jump size distribution, and fractal dimension, our results 27 highlight the profound influence of particle shape on the multiphase flow. 28

²⁹ 1 Introduction

Fluid-fluid displacement in porous media is a common phenomenon encountered 30 in a wide range of natural and industrial processes, such as water infiltration into soil 31 (Lipiec et al., 2006), carbon sequestration (Szulczewski et al., 2012; Matter et al., 2016), 32 enhanced oil recovery (Lake et al., 2014; M. Blunt et al., 1993), and remediation of con-33 tamination in aquifer systems (Nadim et al., 2000). As indicated by the pioneering works 34 by Lenormand et al. (Lenormand et al., 1988; Lenormand & Zarcone, 1989), the multi-35 phase displacement patterns strongly depend on the capillary number (i.e., relative strength 36 of viscous force to capillary force) and the viscosity ratio of the two fluids, and a phase 37 diagram including capillary fingering, viscous fingering, and stable displacement was pre-38 sented. Since then, extensive efforts have been devoted to further investigation of how 39 fluid properties, flow conditions, and topological characteristics of the porous media mod-40 ify the invasion morphology (Yortsos et al., 1997; Armstrong et al., 2014; Rabbani et al., 41 2018; Holtzman, 2016; Wang et al., 2019; Hu et al., 2019; Ju et al., 2020; Xu et al., 2014). 42 Specifically, both numerical and experimental works have revealed the profound influ-43 ence of wettability (i.e., contact angle) in two phase flows (Crisp & Thorpe, 1948; Pur-44 cell, 1950; Mason & Morrow, 1994; Cieplak & Robbins, 1990; Trojer et al., 2015; Jung 45 et al., 2016; Wang et al., 2019, 2020; Holtzman & Segre, 2015; Zhao et al., 2016; Ran et 46 al., 2018; Primkulov et al., 2018). However, the effective contact angle, as one of the key 47 controlling factors, is often unknown prior to the displacement process due to the com-48 plex geometry of pore space. Even for chemically homogeneous porous media, a wide dis-49 tribution of contact angles have been observed due to roughness and pinning of menisci 50 at sharp edges (AlRatrout et al., 2018; M. J. Blunt et al., 2019, 2021). Therefore, it is 51 important to understand how particle shape affects the effective contact angles, which 52 can consequently alter the pore-scale instability events and macroscopic invasion mor-53 phology (Cieplak & Robbins, 1990; Holtzman & Segre, 2015; Geistlinger & Zulfigar, n.d.; 54 Zulfigar et al., n.d.; AlRatrout et al., 2018). 55

In the quasi-static regime of multiphase flow where capillary force dominates the 56 displacement, various numerical approaches have been developed to supplement exper-57 iments, including Navier-Stokes equation solvers and pore-network models. The meth-58 ods of the latter category have been successfully applied in investigation of macroscopic 59 invasion patterns due to significantly less computational cost (M. J. Blunt, 1998, 2001; 60 Cieplak & Robbins, 1988, 1990; Holtzman & Segre, 2015; Holtzman, 2016; Primkulov 61 et al., 2018; Hu et al., 2019). A subclass of pore-network models, the interface tracking 62 algorithm, initially proposed by Cieplak and Robbins (Cieplak & Robbins, 1988, 1990) 63 and recently extended by Primkulov et al. (Primkulov et al., 2018) for consideration of 64 corner flow, has been found successful in reproducing multiphase displacement exper-65 iments in Hele-Shaw cells (Chapuis et al., 2008; Trojer et al., 2015; Holtzman & Segre, 66

2015; Holtzman, 2016; Zhao et al., 2016; Ran et al., 2018). This method captures the 67 pore-scale invasion mechanisms by taking into account the local pore geometry, includ-68 ing the cooperative pore-filling event, which stabilizes the invasion during imbibition (Cieplak 69 & Robbins, 1988, 1990; Holtzman & Segre, 2015). However, up to now, this type of pore-70 network models is applicable to perfectly spherical particles, whereas grains with irreg-71 ular shapes are prevalent in natural systems such as sand packs, and solid walls char-72 acterized by surface with sharp edges due to manufacture limitations are encountered 73 in microfluidics. These non-smooth surfaces can often lead to pinning of menisci dur-74 ing displacement process, which results in effective contact angles deviating from the in-75 trinsic one, altering the capillary resistance at local pore/throat. It is worth noting that 76 surface roughness has also proven influential on the effective contact angle during fluid-77 fluid displacement process (Mehmani et al., 2019; Zulfiqar et al., n.d.; AlRatrout et al., 78 2018; Chen et al., 2018). One option to distinguish the impact from particle shape and 79 roughness on contact angle is whether the effect is transient. Due to surface roughness, 80 the effective contact angle can change as the liquid fills the grooves of the surface, lead-81 ing to a time-dependent behaviour of contact angle. This phenomenon has been exten-82 sively observed in experiments (Mishra et al., 2016; Moulinet & Bartolo, 2007; Sbragaglia 83 et al., 2007; Seo et al., 2018; Papadopoulos et al., 2013), where the effective contact an-84 gle could be described by the Wenzel or Cassie-Baxter model, depending on the wetting 85 states (Wenzel, 1936; Cassie & Baxter, 1944; Marmur, 2003; Gao & Yan, 2009). On the 86 other hand, the pinning of the meniscus at sharp edges (Gibbs, 1961; Oliver et al., 1977), 87 is a thermodynamically stable configuration. In the current study, we thus focus on the 88 latter phenomenon in the quasi-static displacement process. 89

Here, we develop an extended pore-network model (called EPONM) to probe the 90 effect of particle shape on quasi-static fluid-fluid displacement. The model incorporates 91 the explicit determination of basic pore-scale instabilities based on the work of Cieplak 92 and Robbins (Cieplak & Robbins, 1988, 1990). It also includes the volume capacitance 93 model (Måløy et al., 1992; Furuberg et al., 1996), which allows us to capture both the 94 evolution of capillary pressure signal and sizes of Haines jumps. Different from the orig-95 inal algorithm where the volume capacitance is a prescribed constant (Måløy et al., 1992; 96 Furuberg et al., 1996), it is calculated based on local pore geometries without extra as-97 sumptions. More importantly, the sharp edge pinning effect is added to consider the pin-98 ning of the menisci (Gibbs, 1961; Oliver et al., 1977). Our results for different grain shapes 99 indicate that increase in angularity leads to wider distribution of contact angles, which 100 explains the observed greater fluctuations in capillary pressure. Besides, it is found that 101 comparing with more spherical particles, the mean capillary pressure for angular grains 102 is greater in drainage whereas smaller in imbibition. We quantify and analyze the cor-103 relation between grain shape and size distribution of Haines jumps, interfacial length, 104 and fractal dimension across a wide range of wetting conditions. The implications of our 105 findings are then discussed. 106

¹⁰⁷ 2 Extended Pore-Network Model

To model the 2D flow patterns observed in Hele-Shaw cells filled with vertical posts (Zhao et al., 2016; Trojer et al., 2015; Hu et al., 2019; Primkulov et al., 2018) with controlled particle shapes, the porous medium is represented by polygons (instead of circles in past studies) placed on two-dimensional triangular lattice. The invading fluid is injected from the center of the simulation domain. Based on a purely geometrical extension of Young-Dupre equation (Gibbs, 1961; Oliver et al., 1977), the equilibrium state of effective contact angle θ measured within the invading fluid at the triple line follows:

$$\theta_0 \le \theta \le \theta_0 + (180^\circ - \alpha),\tag{1}$$

where θ_0 and α are the intrinsic contact angle and the angle subtended by the two surfaces forming the edge, respectively (Fig. 1(A)). Since in this work the focus is placed

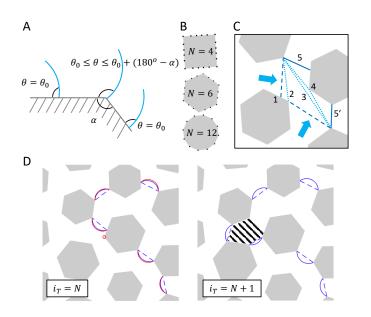


Figure 1. (A) Pinning of menisci at corners leads to greater effective contact angle. (B) Investigated grain shapes in this work with different number of edges N. The black dots represent the mesh points. (C) Schematic showing different invasion types. {1-2, 2-3, 3-4, 4-5(5')} correspond to {unpin, overlap, touch, unpin} events, respectively. Blue arrows mark the movement direction of menisci. (D) Snapshots of invasion morphology at two consecutive steps. Blue solid lines represent menisci after relaxation of previous step. Red solid lines represent menisci at the critical state (in this case an unpin event marked by the red circle will take place). The shaded area is invaded, accompanied by retraction of menisci from red lines at $i_T = N$ to blue lines at $i_T = N + 1$.

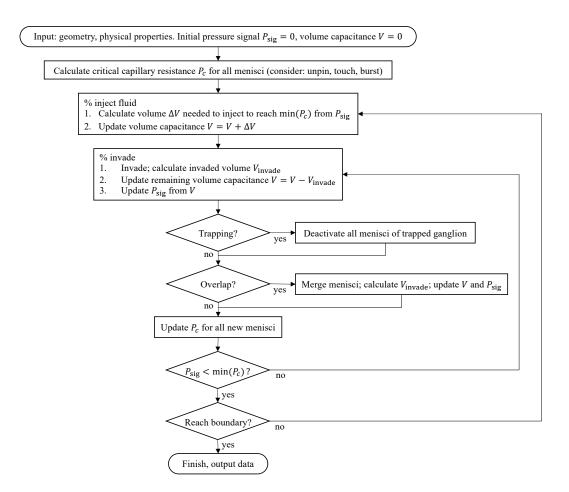


Figure 2. Flow chart of the pore-network model.

on the regime of quasi-static displacement, the advancement of liquid front is governedby capillary force, and the viscous effect is ignored.

In the framework of interface tracking algorithm, the menisci move forward through 119 two types of advancements: (1) pressure-driven events and (2) spontaneous events (re-120 laxation). With a constant injection velocity boundary condition, the capillary pressure 121 builds up accompanied by change in shapes in menisci, until either the meniscus jumps 122 towards the next mesh point due to the local contact angle being greater than the up-123 per bound according to Eqn. 1 (unpin event), or the meniscus touches other grain, form-124 ing two new menisci (touch event). Regarding the fineness of the mesh, the number of 125 mesh points needed per edge is denoted by M = E/N, with E the effective number of 126 mesh points per grain and N the number of edges a grain has. It is found that E = 24127 is sufficient for the studied grain shapes, leading to the corresponding number of mesh 128 points per edge M = 6, 4, 2 for square, hexagon, and dodecagon, respectively (Fig. 1(B)). 129 This is verified by checking both the macroscopic invasion morphology and pore-scale 130 instability events (see Supporting Information for the mesh sensitivity test). After each 131 pressure-driven event, the newly invaded area is subtracted from the total volume ca-132 pacitance (Måløy et al., 1992; Furuberg et al., 1996) (area between red and blue-dashed 133 lines in Fig. 1(D), from which the pressure within the invading fluid is updated accord-134 ing to the remaining volume capacitance, i.e., redistributing the total volume back to each 135 active meniscus assuming all are at thermodynamic equilibrium, i.e., all have the same 136 curvature. Then, potential overlap event and further advancement events including un-137

pin and/or touch are checked and executed. This process is carried out until the small-138 est capillary resistance at the invasion front is greater than the remaining pressure within 139 the invading fluid. Note that after each time step, trapping is checked and all menisci 140 that belong to trapped regions are deactivated to prevent any further movement. Fig. 1(C) 141 shows the schematic of several advancement steps initialized by a pressure-driven un-142 pin event. Fig. 1(D) shows local snapshot of invasion morphology at two consecutive time 143 steps. The blue-solid lines denote the menisci shape after the previous relaxation step, 144 and red-solid lines denote the menisci shape associated with the next minimum critical 145 capillary pressure. The flowchart of the algorithm is shown in Fig. 2. More details on 146 the algorithm, such as the calculation of critical capillary pressure, determination of in-147 stability modes, and conversion between pressure and volume can be found in Support-148 ing Information. 149

To investigate the effect of particle shapes, squares, hexagons, and dodecagons were 150 chosen as representative grains with decreasing angularity (Fig. 1(B)). The 1-by-1 rect-151 angular simulation domain contains in total 7520 particles placed on triangular lattice, 152 corresponding to 94 columns of 80 vertically aligned grains with constant porosity of $0.5912\pm$ 153 0.0005 (see a sample packing structure in Fig. 3(A)). Disorder is introduced by (i) in-154 ducing 10 percent variation in particle size with a uniform distribution and (ii) random 155 rotation of particles in $[0^{\circ}, 360^{\circ})$. The capillary pressure signal, total injected fluid vol-156 ume (area in 2D), and size of Haines jump are recorded at each step, until the invasion 157 front reaches the boundary. Simulations were carried out for each particle shape with 158 five randomly generated porous media under contact angles ranging from 45° to 165° 159 with 15° increment. For displacement processes with contact angles below 45° , the 3D 160 phenomenon corner flow starts to appear(Zhao et al., 2016; Primkulov et al., 2018), which 161 is currently not captured in the model, and thus those processes are not covered in the 162 present work. 163

A typical evolution of the invasion front until breakthrough for hexagonal grains 164 with an intrinsic contact angle of $\theta_0 = 120^\circ$ is shown in Fig. 3(A). Crimson represents 165 the initial stage whereas yellow indicates the late times. The displacement pattern con-166 tains rather ramified structures with significant trapping, which represents the capillary 167 fingering in drainage. Fig. 3(B-i) shows the evolution of the dimensionless capillary pres-168 sure calculated as the curvature $P_c^* = 1/r^*$, with r^* being the radius of the meniscus. 169 The pressure-driven events and spontaneous events are marked by red circles and yel-170 low cross, respectively. Clearly, multiple spontaneous events can take place following a 171 pressure-driven event, which is a manifestation of a Haines jump. In the limit of van-172 ishing capillary number, Haines jump can be regarded as effectively instantaneous com-173 pared with the speed of fluid injection at the inlet. Thus, after conversion from step into 174 time, which is expressed as volume of injected area normalized by the average pore area, 175 Fig. 3(B-ii) shows the pressure signal as a function of A_{inj}^* . The P_c^* values at same in-176 vasion progress are marked by the same number in Fig. 3(B-i) and Fig. 3(B-ii). Specif-177 ically, the processes marked by (1-2) and (2-3) represent a fast Haines jump accompa-178 nied by drop in pressure, and slow injection of invading fluid until the next critical P_c^* 179 is reached, respectively. Fig. 3(B-iii) shows P_c^* signal for the whole simulation. Similar 180 pressure signal signatures in a stick-slip manner have been observed in experiments at 181 quasi-static condition (Måløy et al., 1992; Furuberg et al., 1996; Moura et al., 2020). 182

3 Results and Discussion

The phase diagram of the displacement patterns across a wide range of contact angles for different particle shapes is shown in Fig.4(A). The invasion morphology for medium with dodecagons at $\theta_0 = 45^{\circ}$ is compact with rather smooth front. This hexagonal shape is a direct result of the grain placement in the triangular lattice, which has been observed in previous study (Lenormand, 1990; Holtzman, 2016). However, with increase in angularity, despite the displacement pattern is still relatively stable without trapping, the

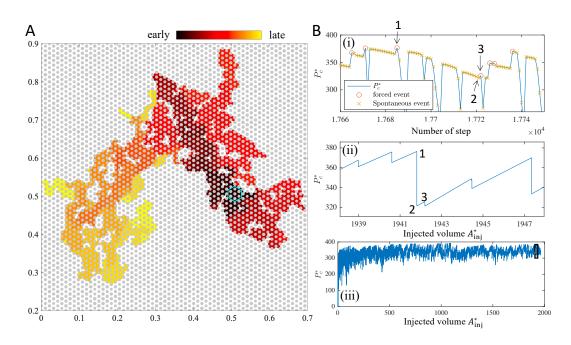


Figure 3. (A) Invasion morphology with hexagon grains (Number of edges N = 6) and $\theta_0 = 120^\circ$. The color represents displacement patterns at different steps. The initial positions of menisci are shown as cyan lines. (B) Process of capillary pressure signal: (i) Evolution of dimensionless capillary pressure calculated as the local curvature $P_c^* = 1/r^*$ in terms of number of step. Red circle marks the critical capillary pressure, where the pressure-driven advancement occurs. Yellow crosses represent the consequent spontaneous events (relaxation). (ii) Conversion from step into time expressed in terms of injected fluid area A_{inj}^* which is normalized by the average pore area. The time for Haines jump is regarded as instantaneous, i.e., only the start and end of P_c^* are "felt" at the inlet. The P_c^* values at same invasion progress are marked by the same number in (i) and (ii). (iii) Capillary pressure signal during the whole simulation. The black box is (ii).

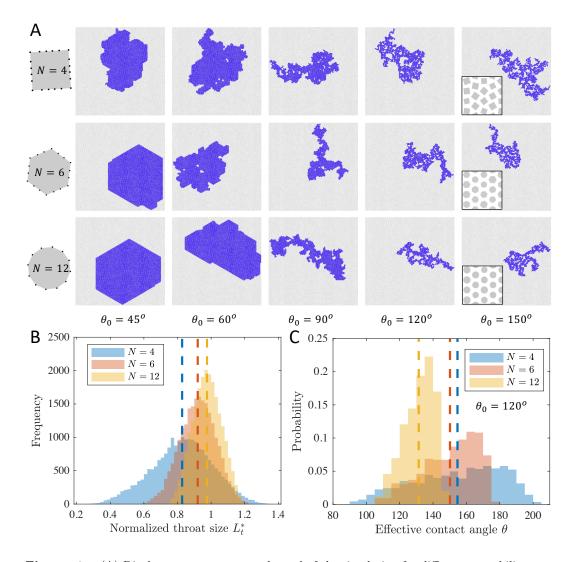


Figure 4. (A) Displacement patterns at the end of the simulation for different wettability conditions and grain shapes. Blue color represents the invading fluid injected from the center of domain. Grey color represents grains. Insets in the last column show the typical grain arrangement of a zoomed region. The simulation ends when the invading fluid reaches the boundary. (B) Distribution of normalized throat size for a typical set of porous media of different grain shapes, calculated as the shortest distance between adjacent grains divided by the throat size of volume equivalent spheres. (C) Contact angle distribution of one typical simulation at the end of simulation with an intrinsic contact angle $\theta_0 = 120^\circ$ for different grain shapes. The dashed lines marks the average value for the corresponding data.

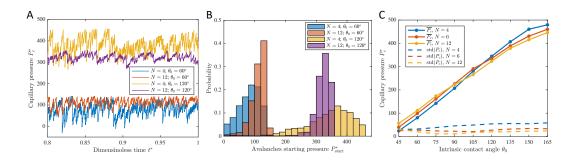


Figure 5. (A) Capillary pressure signals P_c^* for media with squares and dodecagons at $\theta_0 = 60^\circ$ and $\theta_0 = 120^\circ$. Only last 20 percent of invasion is plotted for visualization purpose. (B) Corresponding avalanches starting pressure P_{start}^* distribution for cases in (A), which is the P_c^* at pressure-driven event (red circle in Fig. 3(B-i)). (C) Mean and standard deviation in P_c^* for all grain shapes and wettability conditions. Values are calculated from five individual simulations.

invasion front becomes more irregular, indicating a shift of the dominance of local pore 190 geometry from lattice structure towards grain shape. The results at $\theta_0 = 60^\circ$ demon-191 strate similar trend, with trapping events starting to occur for angular grains. With the 192 increase of θ_0 , the displacement patterns experience a transition from compact displace-193 ment to capillary fingering. The distribution of the normalized throat size L_t^* , calculated 194 as the shortest distance between adjacent grains divided by the throat size of the volume-195 equivalent spheres, is shown in Fig. 4(B). Although the media have similar grain size dis-196 tribution (10 percent variation with uniform distribution) and arrangement (placed on 197 triangular lattice), the L_t^* distribution varies drastically for different grain shapes, with 198 wider span for more angular grains, similar to the effect from increasing topological dis-199 order (Wang et al., 2019). Another influence of particle shape is the smaller average throat 200 size as angularity increases, despite almost constant porosity. For media filled with do-201 decagons, the average throat size is close to 1, i.e., the average throat size is similar to 202 media with perfect spheres, which implies that the shape of dodecagons can be regarded 203 as very close to spherical particles. Fig. 4(C) shows the effective contact angle distribu-204 tion θ at the end of displacement (after the final relaxation process when invasion front 205 reaches the boundary) for the case with an intrinsic contact angle $\theta_0 = 120^\circ$. Note that 206 there is one effective contact angle per triple line (and thus two per meniscus). Due to 207 the sharp edge pinning effect, the distribution narrows for grains with decreasing angu-208 larity. In the case of perfect spheres, one can expect a single value of $\theta = \theta_0$ accord-209 ing to Eqn. (1). Therefore, Fig. 4(B) and Fig. 4(C) summarize the important influences 210 of particle angularity on pore geometry features and contact angle distribution, which 211 will consequently impact the capillary pressure signal and invasion morphology. 212

213

3.1 Capillary Pressure Signal

As indicated in Fig. 5(A), the evolution of dimensionless capillary pressure P_c^* shows 214 larger fluctuations for media with squares compared with more spherical grains (dodecagons), 215 implying greater randomness in local capillary resistance. At the same time, larger mean 216 value is observed with increasing θ_0 , which is a direct result from greater curvature of 217 menisci. The distribution of P_{start}^* , the avalanches starting pressure, can also be directly obtained from the simulation (Fig. 5(B)), where P_{start}^* is the capillary pressure at pressure-218 219 driven event (red circle in Fig. 3(B-i)). For experiments conducted using spherical glass 220 beads in drainage, P_{start}^* is found to distribute within a relatively narrow region (Måløy 221 et al., 1992; Furuberg et al., 1996; Moura et al., 2020). The P_{start}^* distribution is linked 222

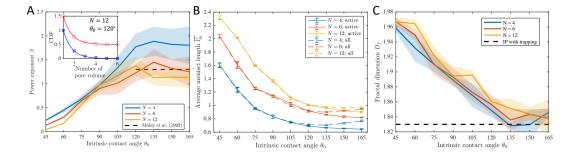


Figure 6. (A) Power exponent β for different grain shapes and wetting conditions. The black-dashed line is from Måløy et al. (Måløy et al., 1992) in drainage experiment with glass beads (since the contact angle is not reported, it is assumed that $\theta_0 > 120^\circ$). Inset: cumulative Haines jump sizes (blue squares) and intervals (red circles) distribution for a typical simulation at N = 12 with $\theta_0 = 120^\circ$. The intervals distribution is shifted by 0.5 for visualization purpose. Lines represent exponential fitting with 1.29 and 1.30 for jump sizes and interval sizes, respectively. (B) The normalized average meniscus width $\overline{l_m^*}$ as a function of intrinsic contact angle. Solid (dashed) lines represent values calculated from active (all) menisci. (C) Fractal dimension calculated using box counting method as a function of intrinsic contact angle, with shaded area showing the standard deviation of five simulations.

to the total volume capacitance stored in all active menisci (Furuberg et al., 1996), which 223 reflects the characteristics of pore geometry. For spherical grains with given packing struc-224 ture, the capillary resistance is distinct between "pore" and "throat", leading to the fact 225 that the avalanches are likely to initialize (and finish) at same location and consequently 226 similar P_{start}^* . As the shape of particle becomes more angular, the distribution of P_{start}^* 227 widens as a result of increased impact from random orientations of grains and pinning 228 of menisci, which is clearly demonstrated in Fig. 5(B). Fig. 5(C) depicts the mean and 229 standard deviation of P_c^* for all grain shapes and wettability with each value calculated 230 from five individual simulations. As expected, the average capillary pressure increases 231 with increasing contact angle, and the fluctuations in capillary pressure are found to be 232 larger for more angular grains across all wetting conditions. Besides, it can be observed 233 that $\overline{P_c^*}$ is greater (smaller) for angular grains in drainage (imbibition) conditions, with 234 a crossover at around $\theta_0 \approx 90^\circ$, which can be attributed to the change in pore and throat 235 size distribution. It is well understood that the throat (pore) size controls the invasion 236 process in drainage (imbibition). Fig. 4(C) indicates that greater angularity leads to smaller 237 average throat size, and in the meantime greater average pore size (as the porosity is the 238 same), which explains the observed higher capillary pressure in drainage and lower cap-239 illary pressure in imbibition. 240

241

3.2 Haines Jumps and Patterns Characteristics

The size of Haines jump can be obtained as the area filled between two pressure-242 driven events (shaded area in Fig. 1(D)), where filling events of single and multiple pores 243 are observed. Both the cumulative pressure jump sizes and intervals distributions dur-244 ing drainage experiments have been found to follow an exponential law (Måløy et al., 245 1992; Furuberg et al., 1996), which is consistent with the simulation results (inset in Fig. 6A). 246 The time interval between two jumps is expressed as the injected area of invading fluid, 247 and the area of invading fluid is expressed in terms of number of average pore volume, 248 calculated as the total pore space divided by the number of pores. Note that the cumu-249 lative intervals distribution is shifted upwards by 0.5 for visualization purpose. The power 250 exponent β , which is regarded as a signature of the displacement process (Måløy et al., 251

1992; Furuberg et al., 1996), is plotted for different grain shapes and wetting conditions 252 (Fig. 6A). It can be seen that β increases with the intrinsic contact angle, reaching a plateau 253 at around $\theta_0 = 120^{\circ}$. Also, in general, the angularity positively correlates with the power 254 exponent. The power exponent β from Måløy et al. (Måløy et al., 1992) in drainage ex-255 periment with glass beads is added as black-dashed line for comparison. Since the con-256 tact angle was not reported, it is assumed that $\theta_0 > 120^\circ$. Their value of β is close to 257 the less angular grains (hexagons and dodecagons), which is consistent with the fact that 258 glass beads are comparatively round and smooth. 259

The average meniscus width $\overline{l_m^*}$ (or the average throat size where menisci are present), 260 normalized by the throat size of porous medium of same porosity filled with mono-dispersed 261 spheres, can reveal the distribution of menisci sizes for different particle shapes and wet-262 ting conditions. Fig. 6(B) shows $\overline{l_m^*}$ as a function of intrinsic contact angle for different 263 angularities with or without consideration of menisci belonging to trapped region. For 264 all active menisci (solid lines), l_m^* decreases with increasing θ_0 , reflecting stronger sta-265 bility of pinned meniscus at small throat. Besides, despite constant porosity for all sim-266 ulations, the average meniscus size is found to be smaller in angular grains as a result 267 of (i) wider distribution of throat sizes (Fig. 4(B)), and (ii) greater capacity of pinning 268 (upper bound in Eqn. (1)) as the local corner becomes sharper, leading to wider distri-269 bution of effective contact angles (Fig. 4(C)). If both active and inactive menisci are con-270 sidered, however, a non-monotonic relationship is observed. This is a result of incom-271 pressibility of the trapped ganglia that prevent the menisci from further advancement 272 and ultimately being pinned at narrower throats. Due to increased amount of trapping 273 at larger contact angle (see Fig. 4(A)), this effect becomes significant at extreme non-274 wetting condition that leads to increase in $\overline{l_m^*}$. This also implies that the pressure within 275 the trapped ganglia could be lower than the capillary pressure signal measured at the 276 inlet. 277

To quantify the displacement patterns, the fractal dimension D_f , as a measurement 278 of the degree to which a pattern fills space, is calculated using box counting method. Fig. 6(C)279 demonstrates the transition from stable displacement with a D_f of around 1.96 towards 280 the regime of capillary fingering with $D_f \approx 1.84$, which is consistent with previously 281 documented values of 1.96 and 1.83 for compact growth and invasion percolation, respec-282 tively (Wilkinson & Willemsen, 1983; Lenormand & Zarcone, 1989; Trojer et al., 2015; 283 Zhao et al., 2016; M. J. Blunt, 2017; Primkulov et al., 2018). Furthermore, in spite of 284 considerable variation (standard deviation, represented by the shaded area), an early tran-285 sition towards capillary fingering, i.e., smaller D_f at the same θ_0 , can be observed for 286 grains with greater angularity, which confirms the qualitative observation in the displace-287 ment patterns in Fig.4(A). This could be partially explained by, apart from the varia-288 tion in local pore structure, the increase of average effective contact angles in angular 289 grains assemblies due to sharp edge pinning effect, which is evident in Fig. 4(C). 290

In this study, though we considered simplified particle shapes (regular polygons), 291 more general and complex shapes can be easily implemented by changing the coordinates 292 of grain vertices and updating the local corner angles accordingly. Also, it is possible to 293 consider the viscous effect by incorporating, for example, the recently proposed moving 294 capacitor model (Primkulov et al., 2019). Furthermore, in the current work the change 295 in distribution of effective contact angle and throat size have been regarded as a direct 296 result of variation in particle shape (Fig. 4(B) and Fig. 4(C)), as simultaneous variations 297 in these two quantities, under fixed porosity, are inevitable with change in particle shape. 298 It could be interesting to solely look at the impact of menisci pinning phenomena at sharp 299 edges by excluding the change in throat (or pore) size distribution, although this would 300 lead to different overall porosity of samples. Note that, for the choice of particle shapes 301 investigated in this study, square grain is chosen as the most angular case instead of tri-302 angle. This is because that with current porosity 0.5912, there will be overlaps of grains, 303 if the shape is triangle, when they are randomly rotated. Since the porosity is already 304

relatively large and we do not want to further increase it, also, we try to avoid manu-305 ally specifying extra criterion during media generation and ensure fully random (homo-306 geneous and isotropic) porous media, the most angular shape for current grain arrange-307 ment and porosity without grain overlap is square. On the other hand, dodecagon is used 308 to represent circular grain, as the algorithm is not capable of doing perfect circles since 309 the calculation of critical capillary pressure and advancement of menisci is based on dis-310 cretization of the grains, and perfect circles correspond to infinite number of mesh points. 311 Nevertheless, it was shown that the case for dodecagon is close to circle, as the average 312 throat size is very close to the porous media filled with perfect circles (Fig. 4(B)). 313

4 Conclusions

In conclusion, we presented an extended pore-network model (EPONM) to probe 315 the effect of grain shapes on quasi-static fluid-fluid displacement in porous media. The 316 model incorporates the mechanisms of pore-scale instabilities (Cieplak & Robbins, 1988, 317 1990), volume capacitance model (Måløy et al., 1992; Furuberg et al., 1996), and sharp 318 edge pinning effect (Gibbs, 1961; Oliver et al., 1977). This allows us to reproduce the 319 multiphase flow patterns across a wide range of wetting conditions for different grain shapes. 320 The algorithm, with further extension, i.e., mainly on geometry description and gener-321 alization on determination of instability modes, should be applicable to porous media 322 with arbitrary grain shape/location, such as reconstructed pore geometry from 2D scan 323 of rocks, offering a rigorous approach for investigation of how topological features mod-324 ify the multiphase displacement in porous media. 325

At the pore scale, increase in grain angularity not only introduces greater hetero-326 geneity in pore geometry, but also amplify the effect of menisci pinning at corners. This 327 is directly reflected by the variations in distributions of throat sizes (Fig. 4(B)) and ef-328 fective contact angles (Fig. 4(C)), which consequently impact both the mean value and 329 fluctuation of the capillary pressure signal (Fig. 5(A)). Macroscopically, an earlier tran-330 sition from stable displacement towards the regime of capillary fingering is observed both 331 qualitatively from the invasion morphology (Fig. 4(A)) and quantitatively as indicated 332 by the fractal dimension (Fig. 6(C)). Various characteristic metrics have been calculated 333 for comparison with past experimental works, including the distribution of avalanches 334 starting pressure, Haines jump size and interval. Reasonable agreement is observed, and 335 impacts of grain shape are discussed. In particular, under the condition of same poros-336 ity for all studied cases, the average size of menisci is found to be smaller in porous me-337 dia with angular grains, showing a tendency of pinning at narrower throats as a result 338 of wider distribution of throat sizes (Fig. 4(B)) and greater pinning strength (Eqn. (1)). 339

Our results have provided independent corroboration of wide distribution of contact angles observed experimentally in mineralogically homogeneous porous media. The profound influences of grain shape are highlighted by systematically analyzing the displacement processes, deepening the understanding of the interplay between pore geometry and wettability. The proposed pore-network model offers an efficient approach for investigation of multiphase flow in natural porous media.

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