Research Article

Maximum Sizes of Fluid-Occupied Pores within Hydrate-Bearing Porous Media Composed of Different Host Particles

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Hydraulic properties of hydrate-bearing sediments are largely affected by the maximum size of pores occupied by fluids. However, effects of host particle properties on the maximum size of fluid-occupied pores within hydrate-bearing sediments remain elusive, and differences in the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores evolving with hydrate saturation have not been well understood. In this study, numerical simulations of grain-coating and pore-filling hydrate nucleation and growth within different artificial porous media are performed to quantify the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores during hydrate formation, and how maximum diameters of fluid-occupied pores change with hydrate saturation is analyzed. Then, theoretical models of geometry factors for incircle and hydraulic diameters are proposed based on fractal theory, and variations of fluid-occupied pore shapes during hydrate formation are discussed. Results show that host particle properties have obvious effects on the intrinsic maximum diameters of fluid-occupied pores and introduce discrepancies in evolutions of the maximum pore diameters during hydrate formation. Pore-filling hydrates reduce the maximum incircle and hydraulic diameters of fluid-occupied pores much more significantly than grain-coating hydrates; however, hydrate pore habits have minor effects on the maximum equivalent diameter reduction. Shapes of fluid-occupied pores change little due to the presence of grain-coating hydrates, but pore-filling hydrates lead to much fibrous shapes of fluid-occupied pores.

1. Introduction

Natural gas hydrates vastly stored in marine sediments along the continental margins have a great potential to become one of global unconventional hydrocarbon energy resources [1–3]. Currently, the exploitation of this potential energy resource is still not economically feasible and requires innovative production methods and techniques [4, 5]. New methods and techniques should be well evaluated by numerical simulators prior to field applications, and results of these numerical evaluations largely depend on proper characterizations of coupled processes and appropriate quantifications of physical properties of hydrate-bearing sediments [6–9].

Hydraulic properties of hydrate-bearing sediments are quite crucial [10–12], and they still represent an ongoing research issue although great effort has been made [13–16].

Hydraulic properties (e.g., saturated water permeability, water retention curve, and relative permeability to water and gas) of hydrate-bearing sediments are inherently governed by pore-scale structures of the solid matrix and pores [17–20]. The pore space occupied by water and/or gas within hydrate-bearing sediments shrinks due to solid hydrate formation, and structures of fluid-occupied pores can be highly diverse due to different hydrate pore habits (e.g., grain-coating and pore-filling) even though hydrate saturations are identical. These diverse pore structures are experimentally

observed, and how they change during hydrate formation or dissociation has been quantified by using varieties of parameters with clear physical significances. Examples include porosity, shape factor, Euler characteristic of individual hydrate cluster, fractal dimensions, pore surface, and pore volume and size [21–24]. Various pore sizes (e.g., the critical, mean, and maximum pore sizes) have been correlated to the hydraulic permeability of porous media, and larger pore sizes generally lead to higher values of the hydraulic permeability [25–27]. Hydraulic properties of porous media are significantly affected by the maximum pore size [27–29], and the maximum pore size is experimentally and theoretically correlated with porosity, permeability, and particle size [30–33]. Grain sizes of marine sediments hosting natural gas hydrates are of a wide range from clays and silts to coarse-grained sands, and sand particle shapes are generally different [34–36]. Effects of host sediments properties (e.g., porosity, grain size, and shape) on the maximum size of fluid-occupied pores within hydrate-bearing sediments remain elusive, although papers have been published to clarify how the maximum size of fluid-occupied pores evolve with hydrate saturation during hydrate formation and dissociation [23, 37].

Pores and fluid channels within most porous media in nature are generally nonspherical and noncircular [38–40]. For these irregularly shaped pores, several definitions of pore diameter are applied to quantify pore sizes. Examples include the equivalent diameter [41], the incircle diameter, and the hydraulic diameter [42]. The equivalent diameter λ_e is a diameter of a circle having an area equal to the pore area. The incircle diameter λ_i is determined by using the maximum ball method [43], and it is widely applied to pore network extractions from porous media [16, 24, 44]. The hydraulic diameter λ_h is defined as $\lambda_h = 4A/P$, where A is the cross-sectional area and P is the wetted perimeter of the cross-section. The hydraulic diameter is commonly used to simplify fluid flow in noncircular tubes and channels as round pipe flow [17, 45, 46]. For fluid-occupied pores within hydrate-bearing sediments, the maximum pore size is expected to decrease with increasing hydrate saturation. However, similarities and differences in the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores evolving with hydrate saturation have not been well understood.

This study is aimed at clarifying the effects of host particle properties on the maximum size of fluid-occupied pores within hydrate-bearing porous media and further the understanding of different maximum pore diameters evolving with hydrate saturation. Grain-coating and pore-filling hydrates are randomly nucleated and grew within different artificial porous media to quantify the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores at selected hydrate saturations, followed by analyses of maximum fluid-occupied pore diameters changing due to the presence of gas hydrates. Then, theoretical models for incircle and hydraulic diameter-related geometry factors are proposed based on the fractal theory, and these proposed models are further applied to provide insights into the hydrate saturation and morphology-dependent pore shape changes during hydrate formation.

2. Methods

Ten square images of artificial porous media shown in Figure 1(a) are generated by using the method of [47] for further numerical simulations of hydrate nucleation and growth in this study. Each of these porous media is constructed by randomly placing black particles with unrestricted overlap into a white square image until the desired porosity has been reached. The porosity is calculated as the ratio of white over total pixel numbers in a square image, and the square image has a side length of 300 pixels. The shape of a solid particle is characterized by using a concept of sphericity which is defined as $S = 2\sqrt{\pi A_s}/P_s$, where A_s is the area and P_s is the perimeter of the solid particle [48]. Particle sphericity values are normally no bigger than 1, and $S = 1$ represents a circular solid particle. In addition, $S = 0.86$ stands for an elliptical solid particle with a major over minor diameter ratio of 2, and $S = 0.76$ for a diameter ratio of 3. Sphericity and size values of solid particles used for porous media constructions are summarized in Figure 1(b), and Figure 1(c) shows the intrinsic porosity and median particle diameter map for those artificial porous media.

Random nucleation and growth of grain-coating and pore-filling hydrates within artificial porous media is numerically simulated by using the method of [37]. These numerical simulations continually calculate minimum distances d_s of fluid-occupied pore pixels to solid particle and hydrate pixels, and fluid-occupied pore pixels are selectively turned into hydrate pixels until the desired hydrate saturation S_h has been reached. Different nucleation and growth preferences represent different hydrate pore habits. Grain-coating hydrate nucleation and growth is simulated by stochastically changing candidate pore pixels with $d_s = 1$ px into hydrate pixels. Pore-filling hydrate nucleation and growth is modeled by preferentially seeding hydrate in candidate pore pixels with the highest d_s , followed by randomly changing hydrate-touched pore pixels into hydrate pixels, and the value of the growth parameter is set to be 0.7 in this study. Each case of numerical simulations is 100 times performed to obtain probabilistically acceptable results. For more details, please refer to our previous paper [37].

The maximum equivalent λ_e , incircle λ_i , and hydraulic λ_h diameters of fluid-occupied pores within hydrate-bearing porous media are quantified from $S_h = 0$ to $S_h = 0.8$ at intervals of $S_h = 0.1$. When a preselected hydrate saturation is reached, fluid-occupied pores are extracted from hydrate-bearing porous media by using the function named “bwlable” with $n = 4$ (i.e., 4 connected pixels) in MATLAB 2016Ra, followed by calculations of the area and perimeter for all the fluid-occupied pores. Calculated values of the area and perimeter are further used to determine values of equivalent λ_e and hydraulic λ_h diameters, and the incircle diameter λ_i is quantified by using minimum distance d_s values. Assuming that there is a rectangular fluid-occupied pore with side lengths of 5 px and 6 px in hydrate-bearing porous media (Figure 2(a)), the equivalent diameter is calculated to be $\lambda_e = 6.2$ px, the incircle diameter $\lambda_i = 5.0$ px, and the hydraulic diameter $\lambda_h = 5.5$ px according to their definitions (Figure 2(b)). Values of the maximum equivalent, incircle,

FIGURE 1: Artificial porous media for further numerical simulations of hydrate nucleation and growth in this study. (a) Square images of artificial porous media constructed by randomly placing and freely overlapping particles. The side length of square images is 300 px. Black color represents solid particles, and white color stands for fluid-occupied pores. (b) Size d and sphericity S of solid particles used for porous media constructions. (c) Intrinsic porosity ϕ_0 of artificial porous media and median diameter d_{50} of solid particles.

and hydraulic diameters of pores within hydrate-free porous media are determined and summarized in Figure 2(c) as intrinsic maximum pore diameters.

3. Results

The maximum equivalent Λ_e , incircle Λ_i , and hydraulic Λ_h diameters of fluid-occupied pores within ten images of porous media containing gas hydrates are shown in Figure 3, and all the pore diameters decrease with increasing hydrate saturation S_h . The average value of intrinsic maximum equivalent diameters Λ_{e0} of fluid-occupied pores within ten porous media (Figure 1(a)) is 154.7 ± 41.1 px with a confidence interval of 95.4% (the same below), and it decreases to 29.7 ± 16.7 px (19.2% of the intrinsic value) and 5.6 ± 1.3 px (3.6% of the intrinsic value) when hydrate saturation S_h is 0.8 for grain-coating (Figure 3(a)) and pore-filling (Figure 3(b)) hydrates, respectively. The average value of intrinsic maximum incircle diameters Λ_{i0} of fluid-occupied pores within ten porous media is 47.8 ± 23.3 px, and it decreases to 28.0 ± 16.7 px (58.6% of the intrinsic

value) for grain-coating hydrates (Figure 3(c)) and 1.0 ± 0.0 px (2.1% of the intrinsic value) for pore-filling hydrates (Figure 3(d)) when $S_h = 0.8$. The average value of intrinsic maximum hydraulic diameters Λ_{h0} of fluid-occupied pores within ten porous media is 19.7 ± 9.1 px, and it decreases to 10.7 ± 3.7 px (54.3% of the intrinsic value) and 4.7 ± 0.6 px (23.9% of the intrinsic value) when $S_h = 0.8$ for grain-coating (Figure 3(e)) and pore-filling (Figure 3(f)) hydrates, respectively. It is obvious that pore-filling hydrates reduce values of the maximum equivalent, incircle, and hydraulic diameters more efficiently than grain-coating hydrates when $S_h = 0.8$, and the maximum hydraulic diameter is the least sensitive to hydrate saturation.

Host particle properties (e.g., intrinsic porosity, particle size, and sphericity) obviously affect the intrinsic maximum equivalent Λ_{e0} , incircle Λ_{i0} , and hydraulic Λ_{h0} diameters of fluid-occupied pores (Figure 2(c)) and introduce diversities (41.1 px for Λ_{e0} , 23.3 px for Λ_{i0} , and 9.1 px for Λ_{h0}) in values of the intrinsic maximum pore diameter. All the intrinsic diversities decrease due to the presence of gas hydrates, and this implies that effects of host particle properties decrease

FIGURE 2: Equivalent, incircle, and hydraulic diameters of fluid-occupied pores within hydrate-bearing and hydrate-free porous media. (a) A pixel map of a rectangular pore occupied by fluids within hydrate-bearing porous media. Light gray pixels stand for the fluid-occupied pore with side lengths of 5 and 6 px, while dark gray pixels represent solid particle and hydrate pixels. Area A of the pore is 30 px^2 , and perimeter P is 22 px . Symbol d_s represents the minimum distances of pore pixels to solid particle and hydrate pixels, and the maximum value $(d_s)_{\max}$ in the rectangular pore is 3 px . (b) Values of equivalent λ_e , incircle λ_i , and hydraulic λ_h diameters for the rectangular pore shown in (a). (c) Values of intrinsic maximum equivalent Λ_{e0} , incircle Λ_{i0} , and hydraulic Λ_{h0} diameters in hydrate-free artificial porous media shown in Figure 1(a).

with increasing hydrate saturation. In addition, pore-filling hydrates (Figures 3(b), 3(d), and 3(f)) have more significant effects on the intrinsic diversity reductions than grain-filling hydrates (Figures 3(a), 3(c), and 3(e)) when $S_h = 0.8$.

In order to obtain further understandings of hydrate saturation and morphology-dependent maximum equivalent, incircle, and hydraulic diameters, Figure 4 shows normalized maximum equivalent Λ_e^* , incircle Λ_i^* , and hydraulic Λ_h^* diameters of fluid-occupied pores within hydrate-bearing porous media. Normalized maximum pore diameters are defined as ratios of maximum pore diameters within hydrate-bearing over hydrate-free porous media. All the values of normalized maximum equivalent diameters Λ_e^* of fluid-occupied pores within porous media containing

grain-coating (Figure 4(a)) and pore-filling (Figure 4(b)) hydrates generally fall into the region between upper $\Lambda_e^* = \sqrt{1 - S_h}$ and lower $\Lambda_e^* = 1 - \sqrt{S_h}$ curves, with Λ_e^* values partially lower than $1 - \sqrt{S_h}$ when hydrate saturation is lower (i.e., $S_h < 0.5$) for grain-coating hydrates and higher (i.e., $S_h > 0.5$) for pore-filling hydrates. These two models are derived based on the assumption that gas hydrates uniformly grow into all pores with different sizes, and for their derivations, refer to [29, 37]. Pore-filling hydrates reduce normalized maximum incircle (Figure 4(d)) and hydraulic (Figure 4(f)) diameters of fluid-occupied pores more significantly than grain-coating hydrates (Figures 4(c) and 4(e)). Normalized maximum incircle and hydraulic diameters of fluid-occupied pores decreasing due to the presence of grain-coating hydrates (Figures 4(c) and 4(e)) can be generally described by using $\Lambda_{i,e}^* = \sqrt{1 - S_h}$. Normalized maximum incircle diameter Λ_i^* of fluid-occupied pores within porous media containing pore-filling hydrates decreases with increasing hydrate saturation as $\Lambda_i^* = (1 - \sqrt{S_h})/2$ when $S_h > 0$ in a general trend (the blue dot curve in Figure 4(d)), and normalized maximum hydraulic diameter can be generally depicted by using $\Lambda_h^* = 1 - \sqrt{S_h}$ (Figure 4(f)).

An empirical model $\Lambda_{e,i,h}^* = 1 - (1 - m)\sqrt{S_h} - mS_h^n$ [23, 37] is applied to fit values of different normalized maximum pore diameters, and values of empirical parameters are summarized in Figure 4(g). The empirical model is an alternative form for the weighted average of theoretical models $1 - \sqrt{S_h}$ for pore-filling hydrates and $\sqrt{1 - S_h}$ for grain-coating hydrates [23]. Values of empirical parameters m and n are largely controlled by hydrate pore habits. For grain-coating hydrates, the normalized maximum equivalent diameter of fluid-occupied pores can be described by setting $m = 0.067$ and $n = 70.6$, normalized maximum incircle diameter by setting $m = 0.71$ and $n = 5.98$, and normalized maximum hydraulic diameter by setting $m = 0.51$ and $n = 19.4$. For pore-filling hydrates, the normalized maximum equivalent diameter of fluid-occupied pores can be described by setting $m = 0.95$ and $n = 0.70$, normalized maximum incircle diameter by setting $m = 0.55$ and $n = 0.10$, and normalized maximum hydraulic diameter by setting $m = 0.50$ and $n = 0.79$.

4. Discussion: Pore Shape Changes due to Hydrate Formation

Fractal theory is widely used to characterize pore-scale structures and investigate various physical (e.g., hydraulic and electrical) properties of porous media [49, 50]. In these investigations, pores within porous media are treated as circles with equivalent areas in the two-dimensional space, and the maximum equivalent diameter Λ_e can be calculated as [50]

$$\Lambda_e = \sqrt{\frac{4}{\pi} \frac{2 - D_f}{D_f} \frac{\phi}{1 - \phi} A_{\text{tot}}}, \quad (1)$$

where A_{tot} is the total area of porous media, ϕ is porosity, and D_f is pore-size fractal dimension [28] which can be determined by using the box-counting method [51].

FIGURE 3: The maximum equivalent Λ_e , incircle Λ_i , and hydraulic Λ_h diameters of fluid-occupied pores within hydrate-bearing porous media evolving with hydrate saturation S_h . The maximum equivalent diameter Λ_e for grain-coating (a) and pore-filling (b) hydrates. The maximum incircle diameter Λ_i for grain-coating (c) and pore-filling (d) hydrates. The maximum hydraulic diameter Λ_h for grain-coating (e) and pore-filling (f) hydrates.

For the case that size of a pore within porous media is quantified by using incircle diameter λ_i (Figure 5(a)), area A_p of the pore can be calculated as

$$A_p = c_i \pi \left(\frac{\lambda_i}{2} \right)^2, \quad (2)$$

and c_i is a geometry factor for incircle diameter. For references, the geometry factor for incircle diameter $c_i = 4/\pi$ for square-shaped pores, $c_i = (3\sqrt{2})/\pi$ for regular triangle-shaped pores, and $c_i = 1$ for circular pores. Then, the maximum incircle diameter Λ_i can be easily calculated as

$$\Lambda_i = \sqrt{\frac{1}{c_i} \frac{4}{\pi} \frac{2 - D_f}{D_f} \frac{\phi}{1 - \phi} A_{\text{tot}}}. \quad (3)$$

If hydraulic diameter λ_h is used to quantify sizes of pores within porous media (Figure 5(a)), area A_p of an individual pore can be calculated as

$$A_p = c_h \pi \left(\frac{\lambda_h}{2} \right)^2, \quad (4)$$

and c_h is a geometry factor for hydraulic diameter. For references, the geometry factor for hydraulic diameter $c_h = 4/\pi$ for square-shaped pores, $c_h = (3\sqrt{3})/\pi$ for regular triangle-shaped pores, and $c_h = 1$ for circular pores. Then, the maximum hydraulic diameter Λ_h can be easily calculated as

$$\Lambda_h = \sqrt{\frac{1}{c_h} \frac{4}{\pi} \frac{2 - D_f}{D_f} \frac{\phi}{1 - \phi} A_{\text{tot}}}. \quad (5)$$

Fitting model	$\Delta^* = 1 - (1 - m)\sqrt{S_h} - mS_h^n$					
Hydrate behaviors	Grain-coating hydrates			Pore-filling hydrates		
Model parameters	m	n	R^2	m	n	R^2
Equivalent diameter	0.067	70.6	0.8817	0.95	0.70	0.8832
Incircle diameter	0.71	5.98	0.9148	0.55	0.10	0.9340
Hydraulic diameter	0.51	19.4	0.7158	0.50	0.79	0.8784

(g)

FIGURE 4: Normalized maximum equivalent Δ_e^* , incircle Δ_i^* , and hydraulic Δ_h^* diameters of fluid-occupied pores within hydrate-bearing porous media evolving with hydrate saturation S_h . Normalized maximum equivalent diameter Δ_e^* for grain-coating (a) and pore-filling (b) hydrates. Normalized maximum incircle diameter Δ_i^* for grain-coating (c) and pore-filling (d) hydrates. The blue solid dot curve in (d) is drawn by $\Delta_i^* = (1 - \sqrt{S_h})/2$ when $S_h > 0$ and $\Delta_i^* = 1$ when $S_h = 0$. Normalized maximum hydraulic diameter Δ_h^* for grain-coating (e) and pore-filling (f) hydrates. (g) A fitting model and its parameter values for descriptions of different pore diameters changing with hydrate saturation. The fitting model is drawn as black dashed curves in (a-f).

If grain-coating hydrates uniformly grow in pores with different sizes and do not alter the shape of fluid-occupied pores (Figure 5(b)), area A_{pc} , incircle λ_{ic} , and hydraulic λ_{hc}

diameters of these pores decrease with increasing hydrate saturation S_h as $A_{pc} = A_{p0}(1 - S_h)$, $\lambda_{ic} = \lambda_{i0}\sqrt{1 - S_h}$, and $\lambda_{hc} = \lambda_{h0}\sqrt{1 - S_h}$, respectively. In these equations, subscript 0

FIGURE 5: Irregularly shaped pores within hydrate-free and hydrate-bearing porous media. Gray color stands for solid particle, white for fluid-occupied pores, and yellow for gas hydrates. Red dashed circles represent incircles quantifying fluid-occupied pores. (a) Irregularly shaped pores with different sizes in hydrate-free porous media. (b) Irregularly shaped pores in porous media containing grain-coating hydrates. Gas hydrates uniformly grow in all pores with different sizes, and the shape of fluid-occupied pores before and after hydrate formation is unchanged. (c) Irregularly shaped pores in porous media containing pore-filling hydrates. Gas hydrates uniformly grow in all pores with different sizes, and the shape of solid hydrates is identical with that of intrinsic pores within porous media.

represents the intrinsic (i.e., hydrate-free) condition and subscript c stands for grain-coating hydrates. Then, it is easy to obtain

$$c_{ic} = c_{i0}, \quad (6)$$

according to Equation (2), and

$$c_{hc} = c_{h0}, \quad (7)$$

based on Equation (4). Normalized geometry factor c_{ic}^* for incircle diameter in porous media containing grain-coating hydrates can be defined as

$$c_{ic}^* = \frac{c_{ic}}{c_{i0}} = 1, \quad (8)$$

according to Equation (6), and normalized geometry factor c_{hc}^* for hydraulic diameter as

$$c_{hc}^* = \frac{c_{hc}}{c_{h0}} = 1, \quad (9)$$

according to Equation (7).

If pore-filling hydrates uniformly grow in pores with different sizes and the hydrate shape is identical with the intrinsic pore shape (Figure 5(c)), area A_{pf} , incircle λ_{if} , and hydraulic λ_{hf} diameters of these pores decrease with increasing hydrate saturation S_h as $A_{pf} = A_{p0}(1 - S_h)$, $\lambda_{if} = \lambda_{i0}(1 - \sqrt{S_h})/2$, and $\lambda_{hf} = \lambda_{i0}(1 - \sqrt{S_h})$, respectively. In these equations, subscript f represents pore-filling hydrates. Then, it is easy to obtain

$$c_{if} = \frac{4(1 + \sqrt{S_h})}{1 - \sqrt{S_h}} c_{i0}, \quad (10)$$

according to Equation (2), and

$$c_{hf} = \frac{1 + \sqrt{S_h}}{1 - \sqrt{S_h}} c_{h0}, \quad (11)$$

based on Equation (4). Equation (10) is applicable for $S_h > 0$, and $c_{if} = c_{i0}$ when $S_h = 0$. Normalized geometry factor c_{if}^* for incircle diameter in porous media containing pore-filling hydrates can be defined as

$$c_{if}^* = \frac{c_{if}}{c_{i0}} = \frac{4(1 + \sqrt{S_h})}{1 - \sqrt{S_h}}, \quad S_h > 0, \quad (12)$$

according to Equation (10), and normalized geometry factor c_{hf}^* for hydraulic diameter as

$$c_{hf}^* = \frac{c_{hf}}{c_{h0}} = \frac{1 + \sqrt{S_h}}{1 - \sqrt{S_h}}, \quad (13)$$

according to Equation (11).

Values of the pore-size fractal dimension for fluid-occupied pores within porous media containing grain-coating and pore-filling hydrates are summarized in Tables 1 and 2, respectively. The pore diameter ratio β defined as the ratio of the minimum over maximum pore diameters can be calculated by [52]

$$\beta = \phi^{1/(2-D_f)}, \quad (14)$$

and values of the pore diameter ratio within porous media containing grain-coating and pore-filling hydrates are shown in Figure 6. It is obvious that all the β values are generally smaller than 1.0×10^{-2} , and the fractal theory can be used to analyze properties of porous media containing gas hydrates in this study [52].

Values of geometry factor c_i for incircle diameter within hydrate-bearing porous media can be calculated by using Equations (3), where $A_{tot} = 300 \times 300 \text{ px}^2$. Geometry factors

TABLE 1: Pore-size fractal dimensions of fluid-occupied pores in porous media containing grain-coating hydrates.

S_h	Porous media number									
	PM-0	PM-1	PM-2	PM-3	PM-4	PM-5	PM-6	PM-7	PM-8	PM-9
0	1.7912 ± 0.0000	1.7777 ± 0.0000	1.7774 ± 0.0000	1.7619 ± 0.0000	1.8171 ± 0.0000	1.7706 ± 0.0000	1.7580 ± 0.0000	1.7916 ± 0.0000	1.7709 ± 0.0000	1.7677 ± 0.0000
0.1	1.7775 ± 0.0001	1.7646 ± 0.0000	1.7652 ± 0.0000	1.7486 ± 0.0001	1.8036 ± 0.0000	1.7563 ± 0.0000	1.7432 ± 0.0000	1.7788 ± 0.0000	1.7569 ± 0.0001	1.7539 ± 0.0000
0.2	1.7614 ± 0.0001	1.7484 ± 0.0001	1.7497 ± 0.0001	1.7329 ± 0.0001	1.7873 ± 0.0001	1.7393 ± 0.0001	1.7256 ± 0.0001	1.7638 ± 0.0001	1.7403 ± 0.0001	1.7375 ± 0.0001
0.3	1.7420 ± 0.0001	1.7289 ± 0.0001	1.7311 ± 0.0001	1.7144 ± 0.0002	1.7681 ± 0.0001	1.7189 ± 0.0001	1.7041 ± 0.0001	1.7461 ± 0.0001	1.7209 ± 0.0001	1.7181 ± 0.0001
0.4	1.7186 ± 0.0001	1.7051 ± 0.0001	1.7084 ± 0.0001	1.6924 ± 0.0002	1.7448 ± 0.0001	1.6938 ± 0.0001	1.6774 ± 0.0001	1.7247 ± 0.0001	1.6973 ± 0.0001	1.6943 ± 0.0001
0.5	1.6896 ± 0.0002	1.6755 ± 0.0001	1.6800 ± 0.0001	1.6654 ± 0.0003	1.7164 ± 0.0001	1.6621 ± 0.0002	1.6438 ± 0.0002	1.6984 ± 0.0001	1.6680 ± 0.0001	1.6647 ± 0.0002
0.6	1.6521 ± 0.0002	1.6373 ± 0.0002	1.6435 ± 0.0002	1.6308 ± 0.0003	1.6797 ± 0.0002	1.6205 ± 0.0002	1.6007 ± 0.0002	1.6647 ± 0.0002	1.6298 ± 0.0002	1.6273 ± 0.0002
0.7	1.6005 ± 0.0003	1.5855 ± 0.0003	1.5936 ± 0.0003	1.5842 ± 0.0004	1.6302 ± 0.0002	1.5634 ± 0.0003	1.5446 ± 0.0003	1.6193 ± 0.0002	1.5779 ± 0.0003	1.5772 ± 0.0003
0.8	1.5227 ± 0.0004	1.5085 ± 0.0005	1.5187 ± 0.0004	1.5142 ± 0.0006	1.5569 ± 0.0003	1.4775 ± 0.0004	1.4666 ± 0.0004	1.5513 ± 0.0003	1.4995 ± 0.0004	1.5042 ± 0.0004

TABLE 2: Pore-size fractal dimensions of fluid-occupied pores in porous media containing pore-filling hydrates.

S_h	Porous media number									
	PM-0	PM-1	PM-2	PM-3	PM-4	PM-5	PM-6	PM-7	PM-8	PM-9
0	1.7912 ± 0.0000	1.7777 ± 0.0000	1.7774 ± 0.0000	1.7619 ± 0.0000	1.8171 ± 0.0000	1.7706 ± 0.0000	1.7580 ± 0.0000	1.7916 ± 0.0000	1.7709 ± 0.0000	1.7677 ± 0.0000
0.1	1.7719 ± 0.0001	1.7591 ± 0.0001	1.7595 ± 0.0001	1.7449 ± 0.0001	1.7990 ± 0.0001	1.7511 ± 0.0001	1.7372 ± 0.0001	1.7765 ± 0.0001	1.7533 ± 0.0001	1.7500 ± 0.0001
0.2	1.7551 ± 0.0001	1.7419 ± 0.0001	1.7427 ± 0.0001	1.7290 ± 0.0002	1.7828 ± 0.0001	1.7318 ± 0.0002	1.7158 ± 0.0002	1.7613 ± 0.0001	1.7364 ± 0.0001	1.7317 ± 0.0002
0.3	1.7386 ± 0.0001	1.7247 ± 0.0002	1.7258 ± 0.0002	1.7137 ± 0.0002	1.7662 ± 0.0001	1.7128 ± 0.0003	1.6946 ± 0.0002	1.7457 ± 0.0002	1.7184 ± 0.0002	1.7127 ± 0.0002
0.4	1.7221 ± 0.0001	1.7073 ± 0.0002	1.7087 ± 0.0003	1.6979 ± 0.0002	1.7491 ± 0.0002	1.6938 ± 0.0003	1.6731 ± 0.0003	1.7297 ± 0.0002	1.7002 ± 0.0002	1.6934 ± 0.0003
0.5	1.7046 ± 0.0002	1.6890 ± 0.0002	1.6907 ± 0.0003	1.6812 ± 0.0002	1.7311 ± 0.0002	1.6743 ± 0.0004	1.6515 ± 0.0004	1.7126 ± 0.0002	1.6813 ± 0.0003	1.6732 ± 0.0004
0.6	1.6847 ± 0.0002	1.6689 ± 0.0003	1.6708 ± 0.0003	1.6610 ± 0.0002	1.7110 ± 0.0002	1.6532 ± 0.0004	1.6283 ± 0.0004	1.6926 ± 0.0003	1.6606 ± 0.0004	1.6518 ± 0.0004
0.7	1.6548 ± 0.0002	1.6405 ± 0.0003	1.6422 ± 0.0003	1.6292 ± 0.0002	1.6819 ± 0.0002	1.6261 ± 0.0004	1.6022 ± 0.0004	1.6615 ± 0.0003	1.6326 ± 0.0003	1.6248 ± 0.0004
0.8	1.6046 ± 0.0003	1.5920 ± 0.0003	1.5939 ± 0.0003	1.5771 ± 0.0003	1.6326 ± 0.0002	1.5790 ± 0.0003	1.5579 ± 0.0004	1.6103 ± 0.0002	1.5843 ± 0.0004	1.5778 ± 0.0004

FIGURE 6: Pore diameter ratio β changing due to the presence of grain-coating (a) and pore-filling (b) hydrates within porous media. Regions colored in light green represent fractal theory applicable conditions (i.e., $\beta < 1.0 \times 10^{-2}$).

FIGURE 7: Geometry factors c_i and normalized geometry factors c_i^* for incircle diameter evolving with hydrate saturation S_h . (a) Geometry factor for incircle diameter c_{ic} in porous media containing grain-coating hydrates. (b) Geometry factor for incircle diameter c_{if} in porous media containing pore-filling hydrates. (c) Normalized geometry factor for incircle diameter c_{ic}^* in porous media containing grain-coating hydrates. (d) Normalized geometry factor for incircle diameter c_{if}^* in porous media containing pore-filling hydrates. The red curve is drawn by $c_{if}^* = 4(1 + \sqrt{S_h})/(1 - \sqrt{S_h})$ when $S_h > 0$, and $c_{if}^* = 1$ at $S_h = 0$.

for incircle diameter changing due to the presence of grain-coating and pore-filling hydrates are shown in Figures 7(a) and 7(b), respectively. It is obvious that hydrate saturation S_h has little effects on c_{ic} values compared with c_{if} values. The normalized geometry factor for incircle diameter changing due to the presence of grain-coating hydrates is shown in Figure 7(c) and pore-filling hydrates in Figure 7(d). It is shown that c_{ic}^* values generally stay close to the horizontal red line $c_{ic}^* = 1$ (Figure 7(c)), and c_{if}^* values go through an evolutionary process from below to above the red curve $c_{if}^* = 4(1 + \sqrt{S_h})/(1 - \sqrt{S_h})$ (Figure 7(d)). These discrepancies

between numerical simulated $c_{ic,f}^*$ data and corresponding theoretical models (i.e., Equations (8) and (12)) are mainly due to differences in hydrate pore habits since grain-coating and pore-filling hydrate growths can hardly follow the uniform and self-similar way (Figure 5) strictly. Based on simulated data, an empirical model is proposed as

$$c_{if}^* = 10^{\alpha S_h}, \quad (15)$$

to depict how c_{if}^* values evolve with hydrate saturation. Equation (15) with $\alpha = 3.7$ can capture the essential physics of pore

FIGURE 8: Geometry factors c_h and normalized geometry factors c_h^* for hydraulic diameter evolving with hydrate saturation S_h . (a) Geometry factor for hydraulic diameter c_{hc} in porous media containing grain-coating hydrates. (b) Geometry factor for hydraulic diameter c_{hf} in porous media containing pore-filling hydrates. (c) Normalized geometry factor for hydraulic diameter c_{hc}^* in porous media containing grain-coating hydrates. (d) Normalized geometry factor for hydraulic diameter c_{hf}^* in porous media containing pore-filling hydrates. The black curve is drawn by $c_{hf}^* = (1 + \sqrt{S_h}) / (1 - \sqrt{S_h})$.

shape changes in hydrate-bearing porous media during hydrate formation, while $\alpha = 2.8$ and $\alpha = 5.1$ set the lower and upper bounds.

Geometry factors c_h and normalized geometry factors c_h^* for hydraulic diameter evolving with hydrate saturation S_h are shown in Figure 8. It is obvious that hydrate saturation and morphology effects on c_i and c_i^* values are similar with those on c_h and c_h^* values. Normalized geometry factor c_{hc}^* for hydraulic diameter in porous media containing grain-coating hydrates changes much more mildly compared with normalized geometry factor c_{hf}^* . The black curve in Figure 8(d) drawn by using Equation (13) agrees well with the general trend of c_{hf}^* values increasing due to the presence of pore-filling hydrates. Based on simulated data, another empirical model is proposed for the normalized geometry factor c_{if}^* prediction, which is

$$c_{hf}^* = 1 + \eta S_h^2, \quad (16)$$

with $\eta = 15$ describing the general trend, $\eta = 3$ setting the lower bound, and $\eta = 50$ setting the upper bound.

In general, grain-coating hydrates barely change while pore-filling hydrates significantly enhance c_{ic}^* and c_{hc}^* values, and perimeters of fluid-occupied pores decrease with an increase in grain-coating hydrate saturations but increase due to the presence of pore-filling hydrates according to their definitions. This implies that shapes of fluid-occupied pores do not obviously change due to the presence of grain-

coating hydrates, but they alter to be more fiber-shaped when pore-filling hydrates occur.

5. Conclusions

This study numerically simulates random nucleation and growth of grain-coating and pore-filling hydrates within artificial two-dimensional porous media to quantify the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores, and how maximum pore diameters evolve due to the presence of gas hydrates is analyzed. Theoretical and empirical models of geometry factors for incircle and hydraulic diameters of fluid-occupied pores are proposed based on fractal theory, and these proposed models together with simulated data are further applied to discuss effects of hydrate saturation and morphology on fluid-occupied pore shapes during hydrate formation. Main conclusions are drawn as follows.

Intrinsic porosity, host particle size, and sphericity not only have obvious effects on the intrinsic maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores within hydrate-free porous media but also lead to discrepancies in the maximum pore diameters even through hydrate saturation and morphology are seemingly identical.

Hydrate pore habits have relatively minor effects on the maximum equivalent diameter of fluid-occupied pores decreasing with increasing hydrate saturation. However, pore-filling hydrates reduce the maximum incircle and hydraulic diameters of fluid-occupied pores much more

significantly than grain-coating hydrates especially when hydrate saturation is higher. Values of normalized maximum equivalent diameter Λ_i^* generally fall in the region limited by upper $\sqrt{1 - S_h}$ and lower $1 - \sqrt{S_h}$ bounds. The upper bound can capture the essential physics of normalized maximum incircle and hydraulic diameter reductions due to the presence of grain-coating hydrates, and the lower bound is generally consistent with values of normalized maximum hydraulic diameter during pore-filling hydrate formation. In addition, values of normalized maximum incircle diameter Λ_i^* during the pore-filling hydrate formation approach $\Lambda_i^* = (1 - \sqrt{S_h})/2$ especially when hydrate saturation is larger. The published empirical model $\Lambda^* = 1 - (1 - m)\sqrt{S_h} - mS_h^n$ can be used to predict normalized maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores during hydrate formation.

Shapes of fluid-occupied pores change little due to the presence of grain-coating hydrates, and corresponding geometry factors for incircle and hydraulic diameters are generally unchanged during hydrate formation. On the contrary, pore-filling hydrates lead to significant variations in fluid-occupied pore shapes (i.e., more fibrous). Normalized geometry factor c_{if}^* for incircle diameter increases with increasing pore-filling hydrate saturation S_h as $c_{if}^* = 10^{3.7S_h}$, and normalized geometry factor c_{hf}^* for hydraulic diameter increases as $c_{hf}^* = 1 + 15S_h^2$. Theoretical models $c_{if}^* = 4(1 + \sqrt{S_h})/(1 - \sqrt{S_h})$ and $c_{hf}^* = (1 + \sqrt{S_h})/(1 - \sqrt{S_h})$ can capture the essential physics of geometry factor enhancements due to pore-filling hydrate formation.

In this study, how the maximum incircle pore diameter changes with increasing hydrate saturation has a potential to benefit two-dimensional pore network modeling of hydraulic properties, and conclusions about the maximum hydraulic diameter are able to facilitate capillary-bundle-based theoretical analyses.

Data Availability

All images of artificial porous media in this study can be downloaded from FigShare at https://figshare.com/articles/Artificial_Porous_Media/11988594.

Additional Points

Key Points. (i) Effects of host particle properties on the maximum equivalent, incircle, and hydraulic diameters of fluid-occupied pores are analyzed. (ii) Similarities and differences in different maximum pore diameters of fluid-occupied pores changing with hydrate saturation are clarified. (iii) Variations of fluid-occupied pore shapes due to the presence of grain-coating and pore-filling hydrates are quantitatively characterized.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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