1
2
3 **Characterization of immiscible fluid displacement processes with various capillary**
4
5 **numbers and viscosity ratios in 3D natural sandstone**
6
7
8
9

10
11 Takeshi Tsuji^{1, 2}, Fei Jiang¹ and Kenneth T. Christensen^{1, 3}
12
13
14
15
16
17

- 18
19 1. International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu
20
21 University, Fukuoka, Japan
22
23
24 2. Department of Earth Resource Engineering, Kyushu University, Fukuoka, Japan
25
26
27 3. Department of Aerospace & Mechanical Engineering, Department of Civil &
28
29 Environmental Engineering & Earth Sciences, University of Notre Dame, Notre
30
31 Dame, Indiana, USA
32
33
34
35
36
37
38
39

40 **Corresponding author:**
41

42 Takeshi Tsuji
43

44
45 Kyushu University
46

47
48 744 Motoooka, Nishiku, Fukuoka 819-0395, Japan
49

50
51 Tel & Fax: +81-92-802-6875
52

53 Email: tsuji@i2cner.kyushu-u.ac.jp
54
55
56
57
58
59
60
61
62

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

Keywords: Multiphase flow; 3D digital rock; lattice Boltzmann method; displacement patterns; pore-filling events; pore heterogeneity and connectivity; natural sandstone

Abstract

To characterize the influence of reservoir conditions upon multiphase flow, we calculated fluid displacements (drainage processes) in 3D pore spaces of Berea sandstone using two-phase lattice Boltzmann (LB) simulations. The results of simulations under various conditions were used to classify the resulting two-phase flow behavior into three typical fluid displacement patterns on the diagram of capillary number (Ca) and viscosity ratio of the two fluids (M). In addition, the saturation of the nonwetting phase was calculated and mapped on the Ca – M diagram. We then characterized dynamic pore-filling events (i.e., Haines jumps) from the pressure variation of the nonwetting phase, and linked this behavior to the occurrence of capillary fingering. The results revealed the onset of capillary fingering in 3D natural rock at a higher Ca than in 2D homogeneous granular models, with the crossover region between typical displacement patterns broader than in the homogeneous granular model. Furthermore, saturation of the nonwetting phase mapped on the Ca – M diagram significantly depends on the rock models. These important differences between two-phase flow in 3D natural rock and in 2D homogeneous models could be due to the heterogeneity of pore geometry in the natural rock and differences in pore connectivity. By quantifying two-phase fluid behavior in the target reservoir rock under various conditions (e.g., saturation mapping on the Ca – M diagram), our approach could provide useful information for investigating suitable reservoir conditions for geo-fluid management (e.g., high CO_2 saturation in CO_2 storage).

1. Introduction

The characteristics of fluid flow in porous media have been the subject of great interest in a wide range of scientific and engineering disciplines. In particular, the displacement of one fluid by another (i.e., two-phase flow) is a process that is central to enhanced oil recovery (EOR), carbon capture and storage (CCS), remediation of non-aqueous phase liquids (NAPL), and geothermal systems. In CCS, the behavior of CO₂ within a reservoir can be characterized as two-phase flow in a porous media system—specifically, the displacement of water (wetting phase) by supercritical CO₂ (nonwetting phase) during drainage displacement (Metz et al., 2005; Michael et al., 2010), and CO₂ displacement by water during the imbibition process. Pore-scale interfacial instabilities, such as Haines jumps (Haines, 1930), snap-off and fingering phenomena, relate to the stability, injectivity, mobility and saturation of CO₂ in the reservoir. Therefore, understanding pore-scale CO₂ flow is crucial to estimating critical CO₂ characteristics in the reservoir, including storage capacity, leakage risk, and storage efficiency (e.g., Yamabe et al., 2015).

The two-phase fluid behavior in porous media is influenced by many reservoir parameters, including the viscosity and density of the fluids, interfacial tension, pore structure, and other porous medium characteristics (e.g., wettability and surface roughness) (e.g., Jiang et al., 2014; Al-Raoush, 2009; Nguyen et al., 2006; Wildenschild et al., 2011), all of which vary from one reservoir to the next. A number of numerical and experimental studies have been conducted to understand the complexities of multiphase fluid flow in porous media (e.g., Ahrenholz et al., 2008; Ahrenholz et al.,

2011; Blunt et al., 2013; Cottin et al., 2010; Al-Housseiny et al., 2012; Homsy, 1987; Lehmann et al., 2008; Parmigiani et al., 2014; Weitz et al., 1987; Berg et al., 2013; Kazemifar et al., 2015a,b; Kitamura et al. 2014; Setiawan et al., 2014). Lenormand et al. (1988) demonstrated that the capillary number (Ca) and the viscosity ratio of a two-phase fluid (M) are the two most important factors (dimensionless values) affecting the displacement behaviors of two-phase flow in 2D micro-models. The capillary number is defined as $Ca = \mu_n V_n / \sigma$, where μ_n , V_n , and σ are the viscosity, velocity of the injected nonwetting fluid, and interfacial tension, respectively. The viscosity ratio of a two-phase fluid is defined as the ratio between the viscosity of the injected nonwetting phase to the viscosity of the defending wetting phase. Lenormand et al. (1988) reported that changes in Ca and M lead to three primary displacement patterns in two-phase flow behavior: stable displacement, viscous fingering, and capillary fingering. The stable displacement pattern, which corresponds to a flat interfacial front, can be observed at simultaneously high Ca and M . At high Ca for $M < 1$, tree-like fingers of the invading fluid, known as viscous fingering, lead to unstable displacement. In this regime, capillary forces are negligible as compared to viscous forces. At low Ca (typically less than 10^{-5}), flow in porous media is dominated by capillary forces, and fluid fingers grow in all directions—even backwards against the bulk injection direction. Such patterns are evidence of capillary fingering; whereby fluid displacement is not continuous due to pore-scale capillary instabilities. When Ca or M takes on an intermediate value, two displacement patterns occur simultaneously, with the resultant pattern termed “crossover” (Ferer et al., 2004; Ferer et al., 2005; Ferer et

al., 2007; Cottin et al., 2010). Owing to the intimate dependence of such patterns on pore-scale properties and local thermodynamic conditions, displacement processes in the crossover domain are not yet fully understood (Yamabe et al., 2015). Furthermore, the quantitative details of the $Ca-M$ diagram as well as crossover region depend on the scale of the system (Løvoll et al., 2004; Toussaint et al., 2005; Toussaint et al., 2012), in addition to the pore structure. In larger systems, the boundaries between fluid displacement patterns are not as sharp and the crossover region is typically broader. Therefore, multi displacement patterns can often occur simultaneously in larger systems.

Many studies in the literature have considered the dynamic behavior of the nonwetting phase, including trapping mechanisms, in simple, and often homogeneous 2D, porous matrices (i.e., periodic granular model; e.g., Wang et al. 2012). However, similar studies in 3D true natural rock pores are lacking. Although recent developments in imaging technology (e.g., X-ray microcomputed tomography; micro-CT) enable observation of dynamic immiscible fluid displacement in natural rock (e.g., Haines jumps; Berg et al., 2013), temporal and spatial resolutions of the measurements of these displacements are often limited. Furthermore, it is difficult to quantify detailed displacement behavior over a broad range of reservoir conditions using only laboratory experiments. Thus, numerical calculations of fluid flow in 3D natural rock pores under a broad range of reservoir-relevant conditions are critical to quantifying pore-scale processes in multiphase flow in a more complete manner. Recently, the digital rock approach has been developed to quantify these processes by combining modern

1
2
3 microscopic imaging with numerical simulation (e.g., Tölke et al., 2002; Ahrenholz et al.,
4
5 2006; Sukop et al., 2008; Raeini et al., 2014; Jiang et al., 2014; Jiang and Tsuji, 2014;
6
7 Yamabe et al., 2015). This approach, undertaken in a high-performance computation
8
9 environment, enables us to calculate porous flow in natural rock samples and estimate
10
11 hydrological properties under a broad range of reservoir conditions (e.g., pressures and
12
13 interfacial tension).
14
15
16
17

18
19 In this study, the 3D natural pore geometry of actual Berea sandstone was
20
21 extracted using micro-CT (Figure 1a), and immiscible fluid displacements (drainage
22
23 processes) in the 3D pore model were calculated under a broad range of reservoir
24
25 conditions using two-phase lattice Boltzmann (LB) simulation (McNamara and Zanetti,
26
27 1988; Pan et al., 2004). We then evaluated the displacement characteristics of the two
28
29 phases and positioned them on the phase diagram for immiscible displacement
30
31 characterized by Ca and M (Lenormand et al., 1988). Although several previous studies
32
33 used LB simulation to reveal the fluid behavior of limited pore throats, this study is the
34
35 first to illuminate the displacement patterns in a natural 3D pore geometry on the
36
37 diagram of Ca and M . We also characterized Haines jumps (Haines, 1930; Måløy et al.,
38
39 1992; Berg et al., 2013; Kazemifar et al., 2015a), which are sudden drops in capillary
40
41 pressure (i.e., differences in pressure between nonwetting and wetting phases) when
42
43 the interface between the two phases moves from a pore throat into a wider pore
44
45 body and thus displaces the wetting phase. The pressure variations causing the Haines
46
47 jumps therefore directly relate to the fluid displacement patterns. We discuss herein
48
49 the influence of pore heterogeneity and three-dimensionality upon the fluid
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

displacement behavior, and attempt to identify suitable conditions in geo-fluid managements (e.g., CO₂ storage) by mapping saturation of the nonwetting phase on the *Ca-M* diagram.

2. Methods

2.1 Digital rock model

To reveal fluid behavior in natural sandstone, a digital rock model was constructed from images of a cylindrical core of Berea sandstone taken using a multi-slice micro-CT scanner (Figure 1a). Berea sandstone is a well-sorted sandstone with a relatively large grain size and consists of quartz, feldspar, carbonates, and clay minerals with a mean grain size of ~250 μm (Øren and Bakke, 2003). The resolution of the 2D scanned images was 3.2 μm. Total data sets were composed of 396 slices with intervals of 5 μm. These 2D pixel images were linearly interpolated to construct a voxel-based 3D volume with a 3-μm grid interval (Figure 1a).

Each pixel in the CT images has a gray value that is assumed to be proportional to the density of the material. To identify and label the pore and grain phases within the images, segmentation was carried out using a single grayscale threshold method to match the porosity of Berea sandstone (~21%). The details of this segmentation process are described in Jiang et al. (2014). The pore size distribution extracted from the digital rock (Figure 1b; Jiang and Tsuji, 2015) demonstrates that the typical pore size is ~20 μm. The wide range of pore size indicates the heterogeneity of the pore space of the Berea sandstone.

2.2 Two-phase lattice Boltzmann (LB) simulation

In this study, the fluid velocity field within the 3D pore spaces was calculated using two-phase LB simulation (Figure 2). LB simulation is a computational fluid dynamics method (McNamara and Zanetti, 1988; Huang et al. 2011). Unlike traditional numerical schemes that solve for the macroscopic variables, this approach is based on the microscopic kinematic equation for the particle distribution function of an assembly of pseudo-molecules. The macroscopic variables are obtained by moment integration of these particle distribution functions. Using the Chapman–Enskog expansion, it is possible to derive the incompressible Navier–Stokes equation in small Knudsen and Mach numbers limits (He & Luo 1997; He et al. 1998; Shan et al. 2006). This approach enables simulation of multiphase flow in complex pore structures and is highly suitable for parallel computing (Aidun and Clausen, 2010).

We used the color gradient model (an optimized version of the Rothman–Keller/Gunstensen (RK) model) in LB simulation (Tölke et al., 2006) to treat the multiphase problem. This approach allowed higher M and lower Ca than the previous LB multiphase extensions. Thus, we were able to calculate the flow behavior under a wide range of conditions. The characteristics of each model in LB simulation are described in Yang and Boek (2013). It should be noted that it is difficult to consider a significant density contrast of two-phase fluids using the present model, and such conditions require the use of other more appropriate models (e.g., Bao and Schaefer, 2013). No-slip boundary conditions were imposed at all solid nodes via a halfway

1 bounceback scheme. The lattice pinning problem in this multiphase model, which
2
3 implies that the interface cannot move when the convection flow is weak, has been
4
5 recognized by several authors (Leclaire et al., 2012; Reis and Dellar, 2011; Latva-Kokko
6
7 and Rothman, 2005). Due to this limitation, we simulate the two-phase flow in a
8
9 relatively high Ca ($10^{-5} \sim 10^{-1}$) regime (Figure 3). A pressure drop was imposed between
10
11 the inlet and outlet, while the boundary on the lateral walls is impermeable.
12
13
14
15
16
17
18

19 To increase the computation efficiency of our LB implementation, we utilized
20
21 graphics processor unit (GPU) parallel computing. The advantages of GPU computing,
22
23 including large memory bandwidth, make it possible to conduct large-scale
24
25 computation on a small-scale GPU-based cluster (Tölke and Krafczyk, 2008). Two-phase
26
27 LB simulation has frequently been applied to study pore-scale flow dynamics, though
28
29 for a rather limited number of pore throats, owing to computational complexity. Our
30
31 GPU implementation for LB simulation allows calculations for large digital rock. As such,
32
33 a much larger number of pore throats can be considered, thus capturing the spatial
34
35 heterogeneity inherent in the actual rock. Using this approach, we succeeded in
36
37 calculating multiphase fluid behavior in a digital rock model of $\sim 1000^3$ grid cells (~ 2 mm
38
39 in physical size with ~ 2 μm resolution; Figure 1c). From absolute permeability
40
41 calculations from single-phase flow simulation for a large number of digital rock
42
43 models with difference domain sizes, we identified that the representative elementary
44
45 volume (REV) of the Berea sandstone we used in this study is ~ 1 mm (unpublished
46
47 data).
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

3. Results and interpretation

The digital rock model for LB simulation should be larger than (or close to) the REV (~1 mm) and take into account the multi-pore nature of Berea sandstone (dominant pore size in our sample = 20 μm ; Figure 1b). In addition, we also need to consider the scaling effect (i.e. scale of rock model) in the characterization of two-phase fluid displacement patterns on the Lenormand diagram (Løvoll et al., 2004; Toussaint et al., 2005; Toussaint et al., 2012). The fluid behavior would be mixed in the large rock model. To clarify the boundary of displacement patterns, therefore, we should focus on displacement patterns in a relatively small digital rock domain. From these reasons, the present simulations were conducted for a digital rock model with dimensions of $400 \times 200 \times 200$ lattice units (400 grid cells along the flow direction). Since the realistic scale of one lattice unit was configured as 3 μm , the volume of this rock model was $1.2 \text{ mm} \times 0.6 \text{ mm} \times 0.6 \text{ mm}$. The size of the rock model is also limited by computational capability, and that reported herein represents the largest possible for two-phase flow calculations at many conditions even with the unique GPU-based simulation approach utilized.

The surfaces of solid grains were assumed to be completely hydrophilic. The lattice units each had a density of 1.0, corresponding to 1000 kg/m^3 for both the wetting and nonwetting phases, because it is considered that the mobility ratio rather than the density ratio plays a key role in our simulations. The time step interval in our simulation is $3.6 \times 10^{-6} \text{ s}$. Since we typically calculate the two-phase fluid behavior for ~3,000,000 time-steps, the fluid behaviors during several seconds were characterized

in this study. The viscosity, injection pressure, and interfacial tension were varied to compute the two-phase fluid displacements under a broad range of reservoir-relevant conditions in the parameter space of Ca and M (Figure 3). As shown in Figure 2, our simulations involved injection of the nonwetting fluid into the digital rock model already saturated with water at 51 different conditions. The conditions in this drainage process are shown on the diagram of $\text{Log } Ca$ – $\text{Log } M$ (dots in Figure 3a). The Ca was calculated based on the average fluid velocity of both phases during the calculation time. At high Ca , breakthrough occurred soon after beginning injection of the nonwetting phase (across 400 grid cells in the flow direction). In several cases of low Ca , however, the injected nonwetting phase moves slowly. In such cases, we finished the numerical simulation before the nonwetting phase arrives at the outlet side of rock model because of high computation cost.

3.1 Classification of displacement patterns

Based on observation of the displacement behavior of the two different fluid phases under various conditions (Figure 2), we classified the behaviors into three displacement scenarios on the diagram of Ca and M (Lenormand et al., 1988) shown in Figure 3a. The animations of each simulation cases are carefully checked to see if there is any typical capillary fingering or viscous fingering displacement behavior. The simulation results shown in Figures 2a and 2b exhibit characteristic features of viscous and capillary fingering, respectively. In the viscous fingering regime (Figure 2a), the thin fingers follow preferential flow paths. In contrast, fingers grow in all directions in the

capillary fingering regime (Figure 2b). The detailed characterization for each displacement pattern is discussed in the following section (3.1.1 - 3.1.3).

To quantify displacement patterns and confirm their classification (Figure 3a), we estimated the time variation of the nonwetting-phase saturation (Figure 4a) as well as its frontal position from the inlet (or the most advanced tip of the interface; Figure 4b). Here saturation refers to the free-phase saturation in the porous media, not the dissolved saturation in the formation water. The frontal position of a finger refers to the distance from the inlet to the most deeply penetrated nonwetting fluid in the rock. Thus, backward movements cannot be evaluated from Figure 4b. The time derivative of the frontal position of the nonwetting phase (Figure 4b) indicates the speed of nonwetting-phase movement from the inlet to the outlet of the rock model. We also plotted the saturation as a function of the frontal position of the nonwetting phase (Figure 5). In Figure 5, we displayed the relationships for each displacement pattern (Figure 5b-5d). The gradient of this relationship corresponds to the degree of saturation in the bulk volume from the inlet to the frontal interface of the nonwetting phase. Therefore, the gradient in Figure 5 represents how effectively the nonwetting phase saturation increased. Our results (Figures 4 and 5) showed that saturation and dynamic movement of the nonwetting phase are significantly influenced by reservoir conditions (on $Ca-M$ diagram).

The saturation of the nonwetting phase was calculated when it reached the outlet of the rock (i.e., crossed 400 grid cells), as shown by the background colors in Figure 3a. If the nonwetting fluid did not arrive at the outlet side, we calculated the fitting curve

of the relationship displayed in Figure 5 and estimated the saturation. Therefore, the saturation displayed in Figure 3 is not at equilibrium; the equilibrium saturation is higher than this saturation.

To classify the displacement pattern from the balance between capillary and viscous force, we calculated the pressure of the nonwetting phase (Figure 6). In particular, we focused on the pressure variation at a pore throat to characterize the pore-filling events. The nonwetting phase pressure was calculated by averaging the pressure inside the nonwetting phase in a surrounding subdomain of the target pore throat (Figures 7 and 8). In Figure 9, we compared the pressure variation at the same pore throat under various conditions. In a capillary fingering regime where capillary force is dominant, the pressure should vary at the pore throat.

Using the information described above, we characterized and classified each displacement mechanism as follows.

3.1.1 Capillary fingering

In the regime of capillary fingering (i.e., low Ca), indicated by red dots in Figure 3a, the movement of the nonwetting fluid was characterized by discontinuous burst-like flow motions (possibly Haines jumps). This movement can be clearly observed as a step-like structure in the time variation of the most advanced tip of the interface, indicated by red lines in Figure 4b. The spatial behavior of the nonwetting phase (Figures 7c-7j) demonstrates that the finger front moves not only in the injection direction (forward movement) but also

1
2
3 against the injection direction toward the inlet (backward movement; i.e., a
4
5 reverse Haines jump). Since the backward movements are key characteristics of
6
7 the capillary fingering regime (Yamabe et al., 2015), we can clearly identify
8
9 capillary fingering displacement from these observations. We can also observe a
10
11 step-like relationship on the cross-plot of saturation and the position of the finger
12
13 front (Figure 5d), suggesting that the saturation of the nonwetting phase increases
14
15 while the most advanced tip of the front remains stationary. This behavior can be
16
17 partially explained by backward movements of the nonwetting phase (i.e., a
18
19 reverse Haines jump; Figures 7h and 7j) and reflects the domination of localized
20
21 pore-scale capillary forces in this fingering regime. The acceleration of saturation
22
23 with time (non-linear relationship) seen in Figure 5d also indicates that the
24
25 backward movement of the nonwetting phase and growth of branching fingers
26
27 increase the saturation behind the finger front.
28
29
30
31
32
33
34
35
36

37 The dominant time variation in nonwetting phase pressure that is associated
38
39 with dynamic pore-filling events (i.e., Haines jumps) is the characteristic feature of
40
41 capillary fingering, therefore the pressure variation is useful information to identify
42
43 the boundary of the capillary fingering regime (Figures 6, 7, 9). The detailed
44
45 interpretation of pressure variation is described in next section (3.2).
46
47
48
49
50
51
52

53 **3.1.2 Viscous fingering**

54
55 In the regime of viscous fingering (i.e., high Ca and low M), indicated by blue
56
57 dots in Figure 3a, the nonwetting phase advances faster than in the capillary
58
59
60
61
62
63
64
65

fingering regime (Figure 4b). This behavior indicates that the permeability (or injectivity) is high for this fingering regime (Jiang et al., 2014). Thin fingers typically follow a number of preferential flow paths in the same direction as injection (Figure 2a), and no backward movement of the nonwetting phase was noted under these conditions (Figure 5c). On the cross-plot of nonwetting phase saturation and frontal position (Figure 5c), the viscous fingering regime is seen to be devoid of the step-like structures observed in the capillary fingering regime. The observed linear relationship (Figure 5c) indicates there was no backward movement of the nonwetting phase in the viscous fingering regime.

The pressure of the nonwetting phase did not vary appreciably during pore-filling events (Figure 9). When the flow velocity is high (high Ca), pore-filling events occur very quickly and smoothly. Besides, the viscous pressure drop becomes dominant in this condition. The overall saturation was not high in this regime (color in Figure 3a). Thus, under viscous fingering conditions, the nonwetting phase did not infiltrate the pore space as effectively as it did under capillary fingering conditions.

3.1.3 Stable displacement

In the 3D natural rock pore, the immiscible fluid does not exhibit ideal stable displacement at conditions of high Ca and high M (Figure 3a), where stable displacement is expected from previous studies (e.g., Zhang et al. 2011). Indeed, at the same condition, we successfully replicated ideal stable displacement (i.e., flat

interfacial front) for a quasi-2D homogeneous periodic pore model (Figures 3b, 10, 11). We further discuss the differences between these two models later.

Figure 5b indicates the relationship between saturation and frontal position in the condition close to stable displacement. Although the relationship should be linear in ideal stable displacement, the behavior in Figure 5b is nonlinear (accelerated), as is the case in capillary fingering. This behavior indicates that capillary fingering still occurred in the condition displayed in Figure 5b and implies that the fluid displacement pattern is significantly influenced by the heterogeneity (and likely the 3D nature) of the pore structure. For conditions close to stable displacement, the saturation of the nonwetting phase was high (Figure 3a), suggesting that the nonwetting fluid effectively infiltrated the pore space.

Using the results of simulations at a broad range of reservoir-relevant conditions, we roughly identified the boundaries of the three primary displacement patterns as a function of Ca and M (Figure 3a) for Berea sandstone. The fluid displacement patterns and saturation map shown in Figure 3a is for the specific Berea sandstone we used in this study, and so it would likely be different for other types of reservoir rock.

To evaluate the influence of pore geometry upon the fluid behavior and nonwetting phase saturation on the diagram, we conducted similar LB simulations for a quasi-2D homogeneous periodic pore model (1024×256 grid cells, and 8 grid cells for thickness; Figures 3b, 10). Many previous studies characterized fluid behavior in similar 2D models. Because of the different dimensions of these two pore models,

furthermore, we could obtain insights for the impact of system dimension upon the fluid behavior. In comparison to the crossover region for 2D homogeneous pore geometries (Zhang et al., 2011), seen in Figure 3, the crossover region in the natural Berea sandstone (gray dots in Figure 3) is much less defined. In particular, the capillary fingering in the Berea sandstone occurs at higher Ca condition than that in the 2D homogeneous model. In addition, the saturation of the nonwetting phase (color in Figure 3) is much different between 3D natural sandstone and the 2D homogeneous model.

3.2 Characterization of pore filling events from pressure variation

As previously described, Haines jumps are displacement processes observed during slow drainage (i.e., rapid invasion process spanning several pore volumes; Haines, 1930), as seen in the capillary fingering regime. The local Reynolds numbers associated with such interfacial jump velocity exceed values associated with Darcy-type viscous flow, and inertial effects may play a central role in these pore-scale dynamic events that govern front migration (e.g., Moebius and Or, 2012). The jumps we identified in our results (Figure 7) can influence the nonwetting fluid saturation (Figure 3), and this phenomenon cannot be captured without considering the inertial term in the Navier–Stokes equation. This is particularly true of backward jumps in the capillary fingering regime, which increase the saturation or storage efficiency of the nonwetting phase (Yamabe et al., 2015). Therefore, characterization of pore-filling events (including inertial phenomena; Ferrari and Lunati, 2014) may be crucial for accurate

estimation of saturation in the reservoir (e.g., to evaluate efficiency of CO₂ storage).

We also characterized the dynamic pore-filling events in Berea sandstone based on the variation in nonwetting fluid pressure (Figures 6-9). When pore-filling events occurred in conditions of low Ca , the nonwetting fluid pressure at the pore throat decreased sharply (Figures 7, 8). Prior to these pore-filling events, a noted pressure accumulation was observed (Figure 7). In this regard, the threshold capillary pressure characterizes the ability of a porous medium saturated with a wetting phase to block the nonwetting phase flow. This threshold capillary pressure P_c^{th} is proportional to the interfacial tension and inversely proportional to the effective radius of the pore throat (e.g., Iglaue et al., 2015). This relationship is represented as $P_c^{th} = \frac{2\sigma\cos\theta}{r_t}$, where σ is the interfacial tension, θ is the contact angle and r_t is the pore throat radius. When the nonwetting phase passes through the throat, the pressure difference between the nonwetting and wetting phase, which corresponds to the capillary pressure P_c , should exceed the threshold value (P_c^{th}). Our results clearly show that the nonwetting phase pressure (or capillary pressure) was locally concentrated at the tip of the penetrating fingers just before filling events in the capillary fingering regime (Figure 8). Thus, the burst-like flow that caused further finger penetration through the pore structure was caused by the instantaneous release of pressure concentration at the finger front. This result suggests that the migration of fingers is governed by local flow dynamics in several pores bodies and throats, rather than the macroscopic pressure gradient associated with injection, in a capillary fingering regime.

To illustrate this process, the pressure variation at the pore throat was observed for sequential pore-filling events (Figure 7a). After the nonwetting phase front moved far from the pore throat, there was no further increase in the pressure at the throat, indicating that the nonwetting phase in the pore throat had attained a local equilibrium. In the conditions of capillary fingering ($\text{Log } Ca = -4.19$, $\text{Log } M = -0.5$; Figure 7), the pressure variation is observed during ~ 4 pore filling events after the nonwetting finger passed through the pore throat, indicating that the spatial length of pressure non-equilibrium is $\sim 400 \mu\text{m}$ along the flow direction. This characteristic length scale is changed due to reservoir conditions (Ca and M). The pressure variations before the equilibrium state at the pore throat (or length scale of pressure non-equilibrium state) were larger under conditions of lower Ca , and the jump events influence a large number of pore bodies and throats (Moebius and Or, 2013). Thus, one must consider rock models that are large enough to capture the properties of equilibrium state (i.e., Armstrong et al., 2015). If simulations are conducted at much lower Ca , the model size should be larger than that of the current model because of the longer spatial length scale of pressure non-equilibrium. Continued increase in the speed of computational hardware will eventually allow such larger model systems to be effectively simulated using the GPU-based approaches described herein.

We also investigated the characteristics of pressure drop at the pore throat due to pore-filling events under various reservoir conditions (Figure 9). These results demonstrate that a large pressure drop is characteristic of a low Ca condition (or the capillary fingering regime). In cases of higher Ca , in which the nonwetting phase

1
2
3 smoothly passed through the pore throat, the pressure of the nonwetting phase
4
5 gradually changed during the pore-filling event (Figure 9). This behavior suggests that
6
7 viscous forces are larger than local capillary forces, meaning that the nonwetting phase
8
9 moves through pore throats without a significant local concentration in pressure under
10
11 viscous fingering conditions. Thus, at higher Ca , the macroscopic pressure gradient
12
13 associated with injection dominates over local, pore-scale pressure gradients (Figure
14
15 6a). Note that the pressure concentration increase and drop were not observed when
16
17 Ca was higher than the condition displayed in Figure 9. The pressure variation of the
18
19 Berea sandstone changed gradually at the boundaries between regimes in the phase
20
21 diagram (Figure 9). Thus, it is difficult to clearly identify the distinct boundaries
22
23 between the primary displacement patterns in the case of 3D heterogeneous natural
24
25 sandstone.
26
27
28
29
30
31
32
33

34
35 The pressure variation further showed that the speed of dynamic pore filling
36
37 events (Reynolds number Re) is locally high with decreasing M or Ca (see time interval
38
39 between two arrows in each panel of Figure 9). This pressure analysis demonstrates
40
41 that the pressure variation (i.e., speed of pore filling events) is directly linked to the
42
43 pore-scale characteristics of the displacement patterns, meaning that it can be used to
44
45 classify the fluid displacement patterns.
46
47
48
49
50
51
52

53 **4. Discussion**

54 **4.1 Influence of pore heterogeneity on the displacement patterns**

55
56
57
58 The boundaries of the three displacement phases on the diagram of Ca and M
59
60
61

(Lenormand et al., 1988) are clearly dependent on the pore structure, particularly the local pore geometry. The present results roughly determined the phase boundaries between the three primary displacement patterns for 3D Berea sandstone (Figure 3a). However, these results are significantly different from the results in the 2D periodic homogeneous pore model (Figure 3b) and results reported in previous studies (e.g., gray dashed lines in Figure 3; Zhang et al., 2011; Huang et al., 2014). In particular, capillary fingering was observed in the natural sandstone in a higher Ca condition ($\text{Log } Ca < -3$; Figure 3a), which was not the case with 2D homogeneous pore geometries (Figure 3b). Furthermore, the crossover region, where two-phase flow behavior includes mixed displacement patterns, was less clearly defined in the case of natural sandstone. Finally, stable displacement was not observed in the 3D natural rock at the condition of high Ca and high M (Figure 3a); although this regime did yield stable displacement within a 2D homogeneous synthetic pore model (Figures 3b, 11). Because of differences in the fluid flow behavior between 3D natural sandstone and 2D homogeneous model, the saturation of the nonwetting phase on the Ca - M diagram is also significantly different (colors in Figure 3a and 3b).

We suggest that these important differences between natural sandstone and homogeneous pore geometries are primarily attributable to the inherent heterogeneity of the natural pore geometry. The natural rock embodies a wide variety of pores (from narrow to wide throats; Figure 1b). Thus high-threshold capillary pressure is required when the nonwetting phase passes through the narrow pore throats. Because the narrow throats are heterogeneously distributed in the natural

rock, the nonwetting phase is therefore heterogeneously trapped at the narrow throats while the pressure perturbation is also dominant (Figure 6). At pores with low pressure gradients, capillary fingering locally occurred. As a result, capillary fingering occurred at relatively high Ca in natural sandstone (Figure 3). And, because multiple displacement patterns occurred simultaneously in the natural sandstone, the crossover region between the displacement regimes was broad and less clearly defined (Figure 3). Because the Berea sandstone used herein is well sorted and less heterogeneous than other natural rocks characteristic of candidate reservoirs, capillary fingering is likely the dominant fluid displacement regime in more heterogeneous (less sorted) natural reservoir rocks.

Apart from pore heterogeneity of the digital rock sample studied herein, another potentially important difference between Lenormand's phase diagram for 2D model and 3D natural sandstone (Figure 3) could be the system's dimensionality. Because the pore connectivity and coordination number in a 3D system is higher than in a 2D system, the variability of pore throats in a 3D model is higher than that of a 2D system. Although it is difficult to quantify the difference between 3D and 2D system from the results presented herein (two cases; Figure 3a and 3b), the dimensional difference could also impact the morphology of the phase diagram.

Finally, differences in the fluid flow patterns noted in the $Ca-M$ diagram could also be associated with the scale of the system considered. In larger systems, the crossover region will likely be broader, making it difficult to identify sharp boundaries in a phase diagram (Toussaint et al., 2012). Both capillary fingering (at smaller scale)

1
2
3 and viscous fingering (at larger scale) will likely occur simultaneously in large systems.
4
5 Furthermore, when the two fluids involved have different viscosities, the viscous
6
7 pressure drop between two points along the fluid interface will typically be different in
8
9 the two fluids. This viscosity contrast will yield a change in the capillary pressure along
10
11 the fluid interface, which could generate a mixture of viscous fingering and capillary
12
13 fingering in large porous systems. While the model size in this work is the largest
14
15 tractable for two-phase flow with currently-available computational resources, it is not
16
17 very large in an absolute sense. Thus, we can only roughly identify displacement
18
19 patterns and the associated boundaries on the phase diagram (Figure 3).
20
21
22
23
24
25
26
27
28

29 **4.2 Suitable condition for effective saturation**

30
31 This study demonstrates that the specific thermodynamic conditions of the
32
33 reservoir can significantly influence the local two-phase flow displacement patterns as
34
35 well as the saturation (Figure 3). Thus, application of this approach to a particular
36
37 reservoir rock will help one identify preferable conditions for optimal reservoir
38
39 management (including injection pressure), such as suitable conditions for geological
40
41 CO₂ storage. Indeed, the saturation of the nonwetting phase on the diagram of Ca and
42
43 M (Figure 3) represents suitable conditions for effective saturation. The saturation of
44
45 the nonwetting phase is significantly altered by the specific reservoir conditions
46
47 considered in this study. Although the boundaries of the three displacement patterns
48
49 were roughly identified herein, the saturation can change gradually or rapidly, even
50
51 within regimes exhibiting the same displacement patterns. Furthermore, our study
52
53
54
55
56
57
58
59
60
61
62
63
64
65

shows that the saturation of the nonwetting phase depends strongly on the details of the rock considered (Figure 3). Thus, quantitative descriptions of the saturation distribution on the Ca - M diagram requires similar simulations for specific target reservoir rocks (3D digital rock). Note that the phase diagram could be also influenced by the system scale, as described above.

In geological CO_2 storage, the saturation and permeability (or injectivity) of CO_2 is of great concern. Although we only report the saturation distribution on the Ca - M diagram in this study, it is also possible to construct permeability distribution on the diagram. When CO_2 is injected into subsurface rocks, the speed of porous flow is slow ($Ca < 1$), and the viscosity ratio between the nonwetting phase (CO_2) and wetting phase (water) is ~ 0.1 (or $\text{Log } M = -1$). The degree of saturation as mapped on the diagram between Ca and M (color in Figure 3) shows that the saturation of CO_2 increases significantly under low Ca . Furthermore, since the viscosity of supercritical CO_2 is more sensitive to changes in reservoir temperature and pressure than the viscosity of brine particularly near the critical point, the flow behavior and the displacement patterns of CO_2 will be strongly tied to the local temperature and pressure of the reservoir. Therefore, in order to maximize CO_2 storage, the injection condition of CO_2 should be carefully selected.

The pore-scale simulation method used herein can be coupled with reservoir simulators by transferring subgrid-scale information to estimate the fluid displacement patterns as well as resulting hydrological properties (e.g., saturation or permeability) at relevant reservoir conditions. At present, field-scale reservoir simulations do not

seriously consider the dynamics of pore-scale fluid displacement patterns, though this study demonstrates that these pore-scale behaviors have a significant impact on the overall saturation and injectivity. In large-scale subsurface reservoirs, different displacement mechanisms will occur simultaneously due to the heterogeneous pore geometry and pressure gradients, and spatially and temporally varying reservoir conditions. In the region close to the fluid injection well, the fluids will displace via viscous fingering because of the high local Ca . However, in most other parts of the reservoir, capillary fingering is likely to be dominant due to a low local Ca . These different dynamic fluid behaviors for each reservoir will undoubtedly influence the security, efficiency, and capacity of fluid injection or extraction projects (e.g., CO_2 geological storage).

5. Summary

The two-phase fluid displacement patterns in 3D natural heterogeneous rock under various reservoir conditions were calculated using LB simulation. The main results of this study include:

- (1) Three typical flow patterns in natural sandstone were classified on the $Ca-M$ diagram. Since we calculated multiphase flow in a large digital rock model containing a number of pore throats, we were able to evaluate the influence of spatial pore heterogeneity inherent in the actual rock. This work demonstrates that the digital rock approach can be effective for revealing pore-scale phenomena.
- (2) Capillary fingering occurred at higher Ca in natural sandstone than in homogeneous

granular model. Stable displacement was not observed in the 3D natural rock at the conditions of high Ca and high M , although this regime did yield stable displacement within a 2D homogeneous pore model. The difference in fluid behavior in 3D natural rock and 2D homogeneous model is likely due to the heterogeneity of the natural pore geometry, including pore connectivity.

- (3) Pressure variations during pore-filling events (Haines jumps) were characterized at several conditions. These variations can be used to diagnose the specific fluid displacement regime. In a capillary fingering regime, pore-flow dynamics in local pore, rather than the macroscopic pressure gradient, govern the migration of fingers. From the pressure variation, we could identify a characteristic length for non-equilibrium state where local pore-flow dynamic is dominant.
- (4) From these results, the most effective or suitable injection conditions for geo-fluid management (e.g., CO_2 storage) in natural sandstone were suggested. Saturation efficiency of the nonwetting fluid could be maximized in capillary fingering and stable displacement regimes. However, the saturation can change gradually or rapidly, even within regimes exhibiting the same displacement patterns. If rock samples are available from a target reservoir, the approach presented herein could be used to explore the fluid displacement patterns in the parameter space of Ca and M , as well as the overall saturation and permeability under such conditions.
- (5) This study shows that several fluid displacement patterns are mixed in the natural reservoir, and this variability in fluid displacement patterns can significantly influence storage capacity associated with geo-fluid management. Therefore,

1
2
3 application of this analysis for the pore geometry of reservoir rock could contribute
4
5 to more effective and accurate fluid management (enhanced saturation in CO₂
6
7 storage).
8
9

10 11 12 13 **Acknowledgements** 14

15
16 We thank Prof. Albert Valocchi (UIUC) for helpful discussion and the anonymous
17
18 reviewers for their many constructive comments, which have greatly improved this
19
20 manuscript. Micro-CT data used to reconstruct the digital rock model is archived by
21
22 JAMSTEC. This study was supported by JSPS through a Grant-in-Aid for Scientific
23
24 Research on Innovative Areas (no. 15H01143), Grant-in-Aid for Scientific Research (A)
25
26 (no. 24246148) and Bilateral Joint Research Projects with MOSR, and JICA/JST through
27
28 SATREPS project. We gratefully acknowledge support of I2CNER, sponsored by the
29
30 World Premier International Research Center Initiative (WPI), MEXT, Japan. KTC
31
32 acknowledges partial support (i.e., partial analysis and interpretation of results) from
33
34 the Center for Geologic Storage of CO₂, an EFRC funded by the U.S. DOE, Office of
35
36 Science, BES, under Award DE-SC0012504.
37
38
39
40
41
42
43
44
45
46
47
48

49 **References** 50

51 Aidun CK, Clausen JR. Lattice-Boltzmann method for complex flows. Ann Rev Fluid
52
53 Mech 2010; 42: 439-72. DOI 10.1146/annurev-fluid-121108-145519.
54
55 Al-Housseiny TT, Tsai PA, Stone HA. Control of interfacial instabilities using flow
56
57 geometry. Nature Phys 2012; 8: 747-50. DOI 10.1038/nphys2396.
58
59
60
61
62
63
64
65

- 1
2
3 Al-Raoush RI. Impact of wettability on pore-scale characteristics of residual
4 nonaqueous phase liquids. *Environmental science & technology* 2009; 43(13):
5 4796-4801.
6
7
8
9 Ahrenholz B, Tölke J, Krafczyk M. Second-order accurate Lattice Boltzmann flow
10 simulations in reconstructed porous media, *International Journal of Computational*
11 *Fluid Dynamics* 2006; 20 (6): 369-377.
12
13
14 Ahrenholz B, Tölke J, Lehmann P, Peters A, Kaestner A, Krafczyk M, Durner W. Prediction
15 of capillary hysteresis in porous material using lattice Boltzmann methods and
16 comparison to experimental data and a morphological pore network model,
17 *Advances in Water Resources* 2008; 31: 1151-1173.
18
19
20
21
22
23 Ahrenholz B, Niessner J, Helmig R, Krafczyk M. Pore-scale determination of parameters
24 for macroscale modeling of evaporation processes in porous media, *Water Resour.*
25 *Res.* 2011; 47, W07543, doi:10.1029/2010WR009519.
26
27
28
29
30 Armstrong RT, Evseev N, Koroteev D, Berg S. Modeling the velocity field during Haines
31 jumps in porous media. *Advances in Water Resources* 2015 (in press).
32 doi:10.1016/j.advwatres.2015.01.008.
33
34
35 Berg S, Ott H, Klapp SA, Schwing A, Neiteler R, Brussee N, Makurat A, Leu L, Enzmann F,
36 Schwarz JO, Kersten M, Irvine S, Stampanoni M. Real-time 3D imaging of Haines
37 jumps in porous media flow. *Proc Natl Acad Sci* 2013; 110(10): 3755-59. DOI
38 10.1073/pnas.1221373110.
39
40
41
42
43 Bao J, Schaefer L. Lattice Boltzmann equation model for multi-component multi-phase
44 flow with high density ratios, *Applied Mathematical Modelling*, 2013; 37(4):
45 1860-1871.
46
47
48
49 Blunt MJ, Bijeljic B, Dong H, Gharbi O, Iglauer S, Mostaghimi P, Paluszny A, Pentland C.
50 Pore-scale imaging and modeling, *Advances in Water Resources* 2013; 51: 197-216.
51 Doi:10.1016/j.advwatres.2012.03.003.
52
53
54
55 Cottin C, Bodiguel H, Colin A. Drainage in two-dimensional porous media: From
56 capillary fingering to viscous flow. *Phys Rev E* 2010; 82: 046315. DOI
57
58
59
60
61
62
63
64
65

10.1103/PhysRevE.82.046315.

Ferer M, Ji C, Bromhal GS, Cook J, Ahmadi G, Smith DH. Crossover from capillary fingering to viscous fingering for immiscible unstable flow: Experiment and modeling. Phys Rev E 2004;70:016303. DOI 10.1103/PhysRevE.70.016303.

Ferer M, Bromhal GS, Smith DH. Two-phase flow in porous media: Crossover from capillary fingering to compact invasion for drainage. Phys Rev E 2005; 71: 026303. DOI 10.1103/PhysRevE.71.026303.

Ferer M, Bromhal GS, Smith DH. Crossover from fractal capillary fingering to compact flow: The effect of stable viscosity ratios. Phys Rev E 2007; 76: 046304. DOI 10.1103/PhysRevE.76.046304.

Ferrari A, Lunati I. Inertial effects during irreversible meniscus reconfiguration in angular pores, Advances in Water Resources 2014; 74, 1-13. DOI 10.1016/j.advwatres.2014.07.009

Homsy GM. Viscous fingering in porous media. Annu Rev Fluid Mech 1987; 19: 271-311. DOI: 10.1146/annur-ev.fl.19.010187.001415.

Haines WB. Studies in the physical properties of soils. V. The hysteresis effect in capillary properties, and the modes of water distribution associated therewith. J Agric Sci 1930; 20 (1): 97-116. DOI: 10.1017/S002185960008864X.

He X, Luo L.-S. Theory of the lattice Boltzmann method: From the Boltzmann equation to the lattice Boltzmann equation. Phys. Rev. E 1997; 56(6): 6811-6817.

He X, Shan X, Doolen GD. Discrete Boltzmann equation model for nonideal gases. Phys. Rev. E, 1998; 57(1): 1-14.

Huang H, Huang J-J, Lu X-Y. Study of immiscible displacements in porous media using a color-gradient based multiphase lattice Boltzmann method, Computer & Fluids 2014; 93: 164-172.

Huang H, Wang L, Lu XY. Evaluation of three lattice Boltzmann models for multiphase flows in porous media. Computers and Mathematics with Applications 2011; 61(12): 3606-17. DOI:10.1016/j.camwa.2010.06.034.

Iglauer S, Pentland CH, Busch A. CO₂ wettability of seal and reservoir rock and the implications for carbon geo-sequestration, *Water Resources Research*, 51, doi:10.1002/2014WR015553.

Jiang F, Tsuji T, Hu C. Elucidating the role of interfacial tension for hydrological properties of two-phase flow in natural sandstone by an improved lattice Boltzmann method. *Transport in Porous Media* 2014; 104(1): 205-29. DOI: 10.1007/s11242-014-0329-0.

Jiang F, Tsuji T. Changes in pore geometry and relative permeability caused by carbonate precipitation in porous media. *Physical Review E* 2014; 90: 053306. DOI: 10.1103/PhysRevE.90.053306.

Jiang F, Tsuji T. Impact of interfacial tension on residual CO₂ clusters in porous sandstone. *Water Resources Research* 2015; 51: 1710-1722, DOI:10.1002/2014WR016070.

Kazemifar F, Blois G, Kyritsis DC, Christensen KT. Quantifying the Flow Dynamics of Supercritical CO₂–Water Displacement in a 2D Porous Micro-Model using Fluorescent Microscopy and Microscopic PIV. *Adv Water Resour* 2015a; DOI:10.1016/j.advwatres.2015.05.011.

Kazemifar F, Blois G, Kyritsis DC, Christensen KT. A Methodology for Velocity Field Measurement in Multiphase High-Pressure Flow of CO₂ and Water in Micromodels. *Water Resour Res* 2015b; 51: 3017-3029.

Kitamura K, Jiang F, Valocchi AJ, Chiyonobu S, Tsuji T, Christensen KT. The study of heterogeneous two-phase flow around small-scale heterogeneity in porous sandstone by measured elastic wave velocities and lattice Boltzmann method simulation. *J Geophys Res* 2014;119:7564-77. DOI :10.1002/2014JB011281.

Latva-Kokko M, Rothman DH. Diffusion properties of gradient-based lattice Boltzmann models of immiscible fluids. *Phys Rev E* 2005; 71: 056702. DOI: 10.1103/PhysRevE.71.056702.

Lake L. *Enhanced Oil Recovery*; Prentice-Hall, Englewood Cliffs, NJ, 1989.

Leclaire S, Reggio M, Trepanier JY. Numerical evaluation of two recoloring operators
 for an immiscible two-phase flow lattice Boltzmann model. *Appl Math Modell* 2012;
 36: 2237-52. DOI:10.1016/j.apm.2011.08.027.

Lehmann P, Berchtold M, Ahrenholz B, Tölke J, Kaestner A, Krafczyk M, Flühler H,
 Künsch HR. Impact of geometrical properties on permeability and fluid phase
 distribution in porous media, *Advances in Water Resources* 2008; 31(9): 1188-1204.

Lenormand R, Touboul E, Zarcone C. Numerical models and experiments on immiscible
 displacements in porous media. *J Fluid Mech* 1988; 189 (9): 165-87.
 DOI:10.1017/S00222112088000953.

Løvoll G, Meheust Y, Toussaint R, Schmittbuhl J, Måløy KJ. Growth activity during
 fingering in a porous Hele-Shaw cell, *Phys. Rev.E* 2004; 70: 026301

Liu H, Valocchi AJ, Werth C, Kang Q, Oostrom M. Pore-scale simulation of liquid CO₂
 displacement of water using a two-phase lattice Boltzmann model. *Advances in*
Water Resources 2014; 73: 144-58. DOI:10.1016/j.advwatres.2014.07.010.

Måløy KJ, Furuberg L, Feder J, Jøssang T. Dynamics of Slow Drainage in Porous Media.
Phys Rev Lett 1992; 68: 2161–4. DOI: 10.1103/PhysRevLett.68.2161.

McNamara G, Zanetti G. Use of the Boltzmann equation to simulate lattice-gas
 automata. *Phys Rev Lett* 1988; 61: 2332.

Metz B, Davidson O, Coninck H, Loos M, Meyer L. IPCC Special Report on Carbon
 Dioxide Capture and Storage. Cambridge University Press: Cambridge, U.K., 2005.

Michael K, Golab A, Shulakova V, Ennis-King J, Allinson G, Sharma S, Aiken T. Geological
 storage of CO₂ in saline aquifers -A review of the experience from existing
 operations. *Int J Greenhouse Gas Control* 2010; 4: 659–67. DOI:
 10.1016/j.ijggc.2009.12.011.

Moebius F, Or D. Interfacial jumps and pressure bursts during fluid displacement in
 interacting irregular capillaries, *J Colloid Interface Sci.* 2012; 377(1):406-415. doi:
 10.1016/j.jcis.2012.03.070

1
 2
 3 Nguyen VH, Sheppard AP, Knackstedt MA, Val Pinczewski W. The effect of
 4
 5 displacement rate on imbibition relative permeability and residual saturation.
 6
 7 Journal of Petroleum Science and Engineering 2006; 52(1–4): 54-70.
 8
 9 Øren PE, Bakke S. Reconstruction of Berea sandstone and pore-scale modeling of
 10
 11 wettability effects. J Pet Sci Eng 2003;39(3–4): 177-99. DOI:
 12
 13 10.1016/S0920-4105(03)00062-7.
 14
 15 Pan C, Hilpert M, Miller CT. Lattice-Boltzmann simulation of two-phase flow in porous
 16
 17 media, Water Resour. Res., 40, W01501, doi:10.1029/2003WR002120.
 18
 19 Parmigiani A, Huber C, Bachmann O. Mush microphysics and the reactivation of crystal
 20
 21 - rich magma reservoirs. J Geophys Res Solid Earth 2014; 119(8): 6308-22.
 22
 23 DOI:10.1002/2014JB011124.
 24
 25 Raeini AQ, Blunt MJ, Bijeljic B. Direct simulations of two-phase flow on micro-CT
 26
 27 images of porous media and upscaling of pore-scale forces. Advances in Water
 28
 29 Resources 2014; 74: 116-26. DOI:10.1016/j.advwatres.2014.08.012.
 30
 31 Reis T, Dellar PJ. A volume-preserving sharpening approach for the propagation of
 32
 33 sharp phase boundaries in multiphase lattice Boltzmann simulations. Comput Fluids
 34
 35 2011; 46(1): 417-21. DOI:10.1016/j.compfluid.2010.12.005.
 36
 37 Setiawan A, Suekane T, Deguchi Y, Kusano K. Three-dimensional imaging of pore-scale
 38
 39 waterflooding phenomena in water-wet and oil-wet porous medium, Journal of Flow
 40
 41 Control, Measurement & Visualization 2014; 2: 25-31.
 42
 43 Shan X, Yuan XF, Chen H. Kinetic theory representation of hydrodynamics: a way
 44
 45 beyond the Navier-Stokes equation. J. Fluid Mech., 550:413–441, 2006.
 46
 47 Sukop MC, Huang H, Li CL, Deo MD, Oh K, Miller DJ. Distribution of Multiphase fluids in
 48
 49 porous media: Comparison between lattice Boltzmann modeling and micro-x-ray
 50
 51 tomography. Phys Rev E 2008; 77: 026710. DOI: 10.1103/PhysRevE.77.026710.
 52
 53 Tölke J, Krafczyk M, Schulz M, Rank E. Lattice Boltzmann simulations of binary flow
 54
 55 through porous media, Phil. Trans. R. Soc. Lond. A, 2002; 360: 535-545.
 56
 57
 58
 59
 60
 61
 62
 63
 64
 65

- Tölke J, Freudiger S, Krafczyk M. An adaptive scheme using hierarchical grids for lattice Boltzmann multi-phase flow simulations. *Computers & Fluids* 2006; 35: 820-830.
- Tölke J, Krafczyk M. Tera FLOP computing on a desktop PC with GPUs for 3D CFD, *International Journal of Computational Fluid Dynamics* 2008; 22: 443-456.
- Toussaint R, Løvoll G, Meheust Y, Schmittbuhl J, Måløy KJ. Influence of pore-scale disorder on viscous fingering during drainage. *Europhys Lett* 2005; 71(4): 583-589.
- Toussaint R, Måløy KJ, Meheust Y, Løvoll G, Jankov M, Sch€ afer G, Schmittbuhl J. Two-phase flow: Structure, upscaling, and consequences for macroscopic transport properties, *Vadose Zone J.* 2012; 11(3), doi: 10.2136/vzj2011.0123.
- Wang Y, Zhang C, Wei N, Oostrom M, Wietsma TW, Li X, Bonneville A. Experimental study of crossover from capillary to viscous fingering for supercritical CO₂–water displacement in a homogeneous pore network, *Environmental Science and Technology* 2012; 47: 212-218
- Weitz DA, Stokes JP, Ball RC, Kushnick AP. Dynamic capillary pressure in porous media: Origin of the viscous-fingering length scale. *Phys Rev Lett* 1987; 59 (26): 2967-70. DOI: 10.1103/PhysRevLett.59.2967.
- Wildenschild D, Armstrong RT, Herring AL, Young IM, Carey JW. Exploring capillary trapping efficiency as a function of interfacial tension, viscosity, and flow rate. *Energy Procedia* 2011; 4(0): 4945-4952.
- Yamabe H, Tsuji T, Liang Y, Matsuoka T. Drainage displacement in porous media: CO₂ saturation and displacement mechanism. *Environ Sci Technol* 2015; 49(1): 537-543.
- Yang J, Boek ES. A comparison study of multi-component lattice Boltzmann models for flow in porous media applications. *Compt Math Appl* 2013; 65(6): 882-890.
- Zhang C, Oostrom M, Grate JW, Wietsma TW, Warner MG. Influence of viscous and capillary forces on immiscible fluid displacement: Pore-scale experimental study in a water-wet micromodel demonstrating viscous and capillary fingering, *Energy & Fuels* 2011; 24: 3493-3505.

Figure Captions

Figure 1. (a) Three-dimensional pore geometry (digital rock model) of the Berea sandstone (400^3 grid cells), reconstructed from micro-CT scanner. (b) Pore size distribution of the Berea sandstone (Jiang and Tsuji, 2015). (c) Example of surface of nonwetting phase during drainage process in the large model (1000^3 grid cells) derived from the LB simulation. In this figure, rock grain (solid part) and wetting phase are transparent.

Figure 2. Surface of the nonwetting phase during drainage process in 3D natural sandstone for each displacement pattern. Nonwetting phase behavior in (a) viscous fingering regime and (b) capillary fingering regime. The left side is the inflow side. These figures were obtained when the front of the nonwetting phase arrived at the outlet side. The model size displayed in this figure ($400 \times 200 \times 200$ grid cells) is used for the LB simulations under various conditions.

Figure 3. Displacement pattern and saturation plotted on the diagram of capillary number Ca and viscosity ratio M . Here we show the results of (a) 3D natural sandstone and (b) 2D homogeneous pore model. The dots indicate the calculation conditions. The nonwetting phase behavior is classified as capillary fingering (red dots), viscous fingering (blue dots) or stable displacement (green dots). The orange dots indicate the capillary fingering close to stable displacement displayed in Figures 4 and 5. The gray

dots indicate the unclassified behavior (crossover state). The red arrows indicate the condition of stable displacement for the 2D homogeneous model (Figure 11b), but not for the 3D natural sandstone (Figure 11a). The color on the phase diagram shows the nonwetting phase saturation when the nonwetting phase arrived at the outlet side (as in Figure 2). The gray-dashed lines are the boundaries of displacement patterns for the 2D homogeneous pore geometry from laboratory experiment (Zhang et al., 2011).

Figure 4. (a) Saturation and (b) frontal position of injected nonwetting fluid as a function of time-step in LB simulation.

Figure 5. The relationship between saturation and the front position of nonwetting phase from the inlet side. In panel (a), we display all relationships. (b) The relationships in the capillary fingering regime close to the stable displacement. The displacement can be characterized as a step-like structure, but the front is always moving along the flow direction. (c) The relationships in the viscous fingering regime. The displacement can be characterized as a smooth curve without a step-like structure. (d) The relationships in the capillary fingering regime. The displacement can be characterized as a step-like structure; saturation increases without moving tip of nonwetting phase.

Figure 6. Pressure distribution of nonwetting phase in viscous fingering and capillary fingering regimes. (a) and (c) Pressure distribution on the 2D slice extracted from 3D digital rock model. Red indicates high pressure of nonwetting phase fluid within the pore space. (b) and (d) Fingers of nonwetting phase surface cross the 2D pressure slice shown in panels (a) and (c).

Figure 7. The pressure and behavior of nonwetting phase during the pore-filling events ($\text{Log } Ca = -4.19$; $\text{Log } M = -0.5$). (a) The time evolution of the average pressure inside the nonwetting phase at the pore throat (yellow arrow in panel b) within the red box in panel (c)–(j). The diameter of the pore throat is $\sim 30 \mu\text{m}$ (see panel b). (b) 2D slice of digital rock perpendicular to the flow direction. Red dots in panel (a) indicate the average pressure measured at each snapshot displayed in panel (c)–(j). The pressure decreases at each pore-filling event. Four pore-filling events are identified from these panels. After the 4th pore filling events, we could not clearly observe pressure drop.

Figure 8. Time variation of nonwetting phase pressure at pore throat during one pore-filling event ($\text{Log } Ca = -4.19$; $\text{Log } M = -0.5$). This pore-filling event corresponds to “event 1” displayed in Figure 7. (a) The average pressure variation of nonwetting phase within the red box in panel (b). Red dots in panel (a) indicate the average pressure measured at each snapshot displayed in panel (c)–(f). The diameter of the pore throat is $\sim 30 \mu\text{m}$, which is the typical size of pore throats in Berea sandstone.

Figure 9. Nonwetting phase pressure variation at the pore throat during the pore-filling events, parameterized by capillary number and viscosity ratio. The location of the pore throat is the same as in Figures 7 and 8. Downward and upward arrows indicate initiation and termination of the pore-filling event, respectively.

Figure 10. The nonwetting phase during drainage process in a 2D homogeneous model for each displacement regime: (a) viscous fingering regime, (b) stable displacement regime and (c) capillary fingering regime. This domain presented is only a portion of the entire 2D homogeneous model domain. Black arrows indicate the injection direction.

Figure 11. Differences in fluid behavior in (a) 3D Berea sandstone and (b) 2D homogeneous granular model at same condition ($\text{Log } Ca = -2$ and $\text{Log } M = 1$). Black arrows indicate the injection direction.

Figure 1

Figure 2

Figure 3

a 3D natural sandstone

b 2D homogeneous model

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

a 3D Berea sandstone
($\text{Log } Ca = -2$, $\text{Log } M = 1$)

b 2D homogeneous granular model
($\text{Log } Ca = -2$, $\text{Log } M = 1$)