Relative Permeability Variation Depending on Viscosity Ratio and Capillary Number

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Abstract

The relative roles of parameters governing relative permeability, a crucial property for two-phase fluid flows, are incompletely known. To characterize the influence of viscosity ratio (M) and capillary number (Ca), we calculated relative permeabilities of nonwetting fluids (knw) and wetting fluids (kw) in a 3D model of Berea sandstone under steady-state condition using the lattice Boltzmann method. We show that knw increases and kw decreases as M increases due to the lubricating effect, locally occurred pore-filling behavior, and instability at fluid interfaces. We also show that knw decreases markedly at low Ca (log Ca < -1.25), whereas kw undergoes negligible change with changing Ca. An M–Ca–knw correlation diagram, displaying the simultaneous effects of M and Ca, shows that they cause knw to vary by an order of magnitude. The color map produced is useful to provide accurate estimates of knw in reservoir-scale simulations and to help identify the optimum properties of the immiscible fluids to be used in a geologic reservoir.

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13	Key Points:
14 15 16 17 18 19	 Relative permeability in two-phase flow is calculated in a three-dimensional digital Berea rock using Lattice Boltzmann Method Relative permeability varies due to lubrication effect, shear force and capillary force, and is related to fluid droplet fragmentation Relative permeability on viscosity ratio-capillary number map is created to predict spatiotemporal variation of reservoir permeability
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31 Abstract

The relative roles of parameters governing relative permeability, a crucial property for two-phase 32 fluid flows, are incompletely known. To characterize the influence of viscosity ratio (M) and 33 capillary number (Ca), we calculated relative permeabilities of nonwetting fluids (k_{nw}) and 34 wetting fluids (k_w) in a 3D model of Berea sandstone under steady-state condition using the 35 lattice Boltzmann method. We show that k_{nw} increases and k_w decreases as M increases due to the 36 37 lubricating effect, locally occurred pore-filling behavior, and instability at fluid interfaces. We also show that k_{nw} decreases markedly at low Ca (log Ca < -1.25), whereas k_w undergoes 38 negligible change with changing Ca. An $M-Ca-k_{nw}$ correlation diagram, displaying the 39 simultaneous effects of M and Ca, shows that they cause k_{nw} to vary by an order of magnitude. 40 The color map produced is useful to provide accurate estimates of k_{nw} in reservoir-scale 41 simulations and to help identify the optimum properties of the immiscible fluids to be used in a 42 geologic reservoir. 43

44 Plain Language Summary

The relative permeability is a crucial parameter in a system where two fluid phases exist 45 simultaneously. For example, in carbon capture and storage, relative permeability is important to 46 47 assess the replacement mechanism of the existing fluid in the reservoir (wetting fluid) by the injected CO_2 (non-wetting fluid). It is also an important parameter in enhanced oil recovery 48 fields, as high relative permeability of oil indicates that the oil in the reservoir can be extracted 49 50 quickly. The relative permeability is temporally and spatially varied by reservoir conditions (e.g., temperature). But currently, in reservoir-scale fluid flow simulation, relative permeability is 51 assumed to be constant regardless of the different conditions. In this study, we conducted 52

simulations to calculate relative permeability in various viscosity ratio (*M*) and capillary number (*Ca*) conditions. We found that relative permeability changes dramatically in different *M* and *Ca* conditions, and we further mapped relative permeability on the diagram between *M* and *Ca* to predict relative permeability accurately in various reservoir conditions. Our findings can be useful to determine the suitable fluid properties to be used in reservoir management and to accurately estimate fluid behavior based on reservoir-scale simulation with variant relative permeability.

60 **1 Introduction**

61 The relative permeability of the different fluids in a two-phase flow has been extensively studied in scientific and engineering fields concerned with two-phase flows in geological 62 reservoirs, such as in carbon capture and storage (CCS) fields, enhanced oil recovery (EOR) 63 operations, geothermal power systems, and geological radioactive waste disposal repositories 64 (Benson et al., 2015; C. Chen & Zhang, 2010; Gudjonsdottir et al., 2015; Niibori et al., 2011; 65 Shad et al., 2008; Wu & Wang, 2020). Relative permeability is a crucial hydraulic property for 66 modeling the flow of both fluids and assessing the mechanisms of fluid displacement in the 67 reservoir. For example, when CO_2 is injected into a CCS reservoir, the CO_2 (nonwetting phase) 68 displaces the existing fluid in the reservoir, such as oil or brine (wetting phase). Relative 69 permeability values can be used to estimate the reduction in CO₂ fluid flow due to surface-70 tension effects between CO_2 and the brine, thus the parameter is useful to assess the injectivity of 71 72 the CO_2 (Benson et al., 2015; Burnside & Naylor, 2014). The relative permeability can also aid estimation of how much fluid can be displaced by CO₂ before the system reaches the wetting 73 fluid irreducible saturation condition, limiting the CO₂ volume that can be stored in the reservoir 74

75	(Burnside & Naylor, 2014). Conversely, in EOR systems, the best results are obtained when the
76	relative permeability is high for oil and low for the injected fluid (Heins et al., 2014).
77	Several factors influence relative permeability in a two-phase flow system. For example,
78	in a CCS project, the fluid relative permeability is affected by the heterogeneity of the rock, such
79	as pore size and pore connectivity (Benson et al., 2015; Fei Jiang & Tsuji, 2014; Zhang et al.,
80	2022). In addition, relative permeability is affected by the interactions between the two fluids,
81	such as interfacial tension, the viscosity and velocity of the fluids, and their wettability (Lefebvre
82	du Prey, 1973). Because fluid viscosity varies dramatically with pressure and temperature, such
83	reservoir environments can further affect relative permeability. Therefore, in a two-phase flow
84	system, the relative permeability value of each fluid is not only a function of saturation; it is also
85	affected by other parameters related to environmental and by the interaction between the two
86	component fluids.

Lenormand et al. (1988) reported that two parameters, viscosity ratio and capillary
number, can explain the interaction between two immiscible fluids. The viscosity ratio (*M*) is a
dimensionless parameter describing the ratio between the viscosity of the injected nonwetting
fluid and the viscosity of the ambient wetting fluid:

91
$$M = \frac{\mu_{\rm nw}}{\mu_{\rm w}} \qquad (1)$$

92 where μ_{nw} is the dynamic viscosity of the nonwetting fluid and μ_w is the dynamic viscosity of the 93 wetting fluid.

94 The capillary number (*Ca*) is a dimensionless parameter describing the ratio between the
95 viscous drag forces and the interfacial tension forces between two immiscible fluids:

96
$$Ca = \frac{\mu_{\rm nw} V_{\rm nw}}{\sigma} \qquad (2)$$

97 where V_{nw} is the average fluid velocity of the nonwetting fluid and σ is the interfacial tension 98 (IFT) between the two fluids.

Despite the recognition that the relative permeability k is a function of M, several studies 99 100 of this relationship have reported divergent results. An experimental study (Odeh, 1959) found that the relative permeability of the nonwetting fluid (k_{nw}) increases and the relative permeability 101 of the wetting fluid (k_w) stays relatively constant as M increases, as have several other 102 experimental and numerical studies (Dou & Zhou, 2013; Goldsmith & Mason, 1963; Huang & 103 104 Lu, 2009; Jeong et al., 2017; Mahmoudi et al., 2017; Yiotis et al., 2007; Zhao et al., 2017). An analytical study of co-current annular flow in which the wetting fluid is distributed on the pore 105 surface and the nonwetting fluid is in the middle of the pore produced empirical equations for the 106 nonwetting and wetting fluids as a function of *M* and saturation: 107

108
$$k_{\rm nw} = S_{\rm nw} \left[\frac{3}{2}M + S_{\rm nw}^2 (1 - \frac{3}{2}M)\right] \quad (3)$$

109
$$k_{\rm w} = \frac{1}{2} (1 - S_{\rm w})^2 (3 - S_{\rm w}) \quad (4)$$

where S_{nw} is the saturation of the nonwetting fluid, and $S_w = 1 - S_{nw}$ is the saturation of the wetting fluid. These equations suggest that k_w is not affected by increasing M and is a function of saturation alone. However, other studies have reported that k_{nw} increases and k_w decreases as Mincreases (Ahmadlouydarab et al., 2012; Fan et al., 2019; Goel et al., 2016; Ramstad et al., 2010); thus, there is as yet no general agreement on the variation of k_w with increasing M. One of the challenges of previous studies was the difficulty of removing the effects of capillary forces and wettability factors when evaluating this relationship.

117	Similarly, studies of the influence of <i>Ca</i> on relative permeability is still incompletely
118	known. In an experimental study, Fulcher et al. (1985) concluded that k_{nw} is a function of IFT
119	and viscosity variables individually rather than a function of the Ca , whereas k_w can be modeled
120	directly as a function of Ca. Several studies (Asar & Handy, 1989; Fan et al., 2019; Harbert,
121	1983) also found that both k_{nw} and k_w increase as IFT decreases because the two fluids interfere
122	less with each other and thus tend to form more well-connected flow pathways. One of the
123	studies (Asar & Handy, 1989) showed that both fluids relative permeability curves tend to
124	straighten and approach the 45° tangent line as IFT approaches zero. Other studies (Amaefule &
125	Handy, 1982; Fei Jiang et al., 2014; Shen et al., 2010) also concluded that both k_{nw} and k_w
126	decrease as Ca decreases. On the other hand, a numerical study (Zhao et al., 2017) concluded
127	that k_w increases with increasing <i>Ca</i> under neutral wetting conditions ($\theta = 90^\circ$) but stays
128	relatively constant with increasing <i>Ca</i> under strong wetting conditions ($\theta = 135^{\circ}$). These
129	divergent results warrant further investigations of how relative permeability changes with
130	changing IFT and Ca. One of the challenges in this evaluation is the difficulty of isolating the
131	effect of IFT while keeping other parameters, such as viscosity, constant. In addition, Ca is a
132	function of fluid velocity, which is a direct of from the simulation. Thus, it is difficult to hold Ca
133	constant in all simulation conditions.

Previous studies have also demonstrated how the viscosity ratio can affect the breakup of fluid droplets. Instabilities due to the viscosity gradient at the interface of the two fluids cause fluid droplet deformation and breakup under viscous flow conditions when the Reynolds number is low (Bischofberger et al., 2015; Mu'min et al., 2021; Nekouei & Vanapalli, 2017; Stone, 138 1994). We suspect that the fluid droplet fragmentation mechanism is also one of the driving factors of the relative permeability change caused by the viscosity ratio. However, to the best of

140	our knowledge, no research linking relative permeability variation to fluid droplet breakup
141	caused by viscosity ratio variation has ever been done.

Previous studies have usually evaluated the separate effects of *M* and *Ca* on relative permeability. However, in two-phase flows, the effects of viscosity gradient and interfacial tension must be evaluated simultaneously to accurately predict the hydraulic properties in the system (Tsuji et al., 2016), including relative permeability. Because few studies have evaluated the condition when both parameters influence relative permeability using a steady-state simulation, our aim in this work was to fill that knowledge gap.

In this paper, we propose a method to simultaneously evaluate the effects of viscosity 148 ratio M and capillary number Ca on relative permeability by creating a $M-Ca-k_{nw}$ relationship 149 map (i.e., showing relative permeability on a diagram relating M and Ca) by applying a color 150 gradient Lattice Boltzmann method (LBM) simulation to a three-dimensional (3D) digital rock 151 152 model. We begin by evaluating the effects of M and Ca individually on the relative permeabilities of the nonwetting and wetting fluids by holding other parameters constant. We 153 also calculate the number of fluid clusters to describe the fluid connectedness, a factor that is 154 155 directly related to relative permeability, and link the fluid connectedness to fluid droplet breakup. We then map k_{nw} for various *M*–*Ca* conditions in search of general trends. We believe that such a 156 map has never before been created, and that it will be useful for quickly estimating relative 157 158 permeability if M and Ca are known. By understanding the variations of relative permeability in 159 the M-Ca parameter space, we can conduct accurate large-scale reservoir simulations by considering the reservoir conditions M and Ca and thereby contribute to the wide range of 160 161 research regarding applications that employ two-phase fluid mixtures.

162 **2 Methods**

163 2.1 Lattice Boltzmann Method

164	The LBM is a branch of computational fluid dynamics that has emerged as a popular
165	technique to solve multiphase fluid flow systems in complex geometries because of its
166	algorithmic simplicity (S. Chen & Doolen, 1998; Dou & Zhou, 2013; Fei Jiang et al., 2014;
167	Ramstad et al., 2010; Succi et al., 2010). LBM simulations treat fluids as a group consisting of
168	fictive particles; the movement of these particles is simulated with a statistical approach. The
169	movement of the bulk fluid is simulated from the propagation and collision processes of the
170	fictive particles (Huang et al., 2011). We chose the Rothman-Keller color gradient model (Tölke
171	et al., 2006) to conduct the simulations because it can simulate a high fluid viscosity ratio with
172	better accuracy than other LBM models (Ahrenholz et al., 2008; Yang & Boek, 2013). The color
173	gradient model is so named because it graphically represents two-phase fluids as a mixture of a
174	wetting and nonwetting fluid, assigned the colors blue and red, respectively (Huang et al., 2015;
175	Fei Jiang et al., 2022). Our 3D simulations used the D3Q19 (3 dimensions, 19 velocity lattice)
176	velocity model.



Fig. 1. Berea sandstone digital rock model: (a) core form and (b) mirrored form.

177

We used a 3D digital rock model obtained from microtomographic images of Berea sandstone (Fig. 1a). Berea sandstone is chosen due to its relatively well-known properties (Øren & Bakke, 2003). The rock has a relatively large mean grain size of ~250 μ m and consists of quartz, feldspar, carbonates, and clay minerals. Its average pore size is ~20 μ m with an average throat size of ~10 μ m. Our rock model, obtained from the database of Dong and Blunt (2009), had an original resolution of 5.345 μ m per pixel. To ensure that the digital rock resolution would be fine enough for the two-phase flow simulation conditions, each pixel was split into 2; hence,

^{178 2.2} Digital rock model

each pixel in our model represents 2.673 μ m. The 3D digital rock is a stack of 400 tomographic images, and each image includes 400 × 400 pixels. Thus, the digital rock represents an actual size of 1.069 mm × 1.069 mm × 1.069 mm. The simulation requires a rock model that is larger than its representative elementary volume (~1 mm for Berea sandstone), and the multi-pore nature of the rock must be considered.

191 2.3 Relative permeability calculation

192 The viscosity ratio *M* is defined by dividing the nonwetting fluid's viscosity (μ_{nw}) by the 193 wetting fluid's viscosity (μ_w) (equation 1). In the LBM, μ can be calculated as follows:

194
$$\mu = \frac{1}{3}(\tau - \frac{1}{2})$$
 (5)

where τ represents the relaxation parameter. Thus, the viscosities of both fluids can be modified 195 by changing the τ value. Because τ must be larger than 0.5 for a positive viscosity and the 196 simulation becomes unstable as τ approaches 0.5, it is important that the simulation uses a τ 197 value that ensures its stability and accuracy. In this study, we altered M only by changing the τ 198 value of the nonwetting fluid to manipulate μ_{nw} . The wetting fluid viscosity was kept constant at 199 0.1555 lu²/ts (length²/time in lattice units) in all simulations to preserve its stability. Changes in 200 201 *M* can be interpreted as a change in either μ_w or μ_{nw} ; thus, modifying either viscosity value will 202 produce the same results if their ratio is maintained.

The capillary number *Ca* can be modified based on equation (2) by changing the σ and μ_{nw} parameters. Because μ_{nw} is also used to determine *M*, we achieved the desired *Ca* by adjusting σ alone. In the color gradient LBM model, σ can be modeled by introducing a perturbation term into equilibrium equations based on the gradient of the phase field of the two

fluids. Tölke et al. (2006) provides a detailed description of the model. The average velocity of the nonwetting fluid after the simulation had converged to equilibrium was used to calculate the *Ca* number. We adjusted σ at every simulation point (every *M* and in every saturation condition) to ensure that *Ca* had a similar value and fell within the permitted error range of log *Ca* ± 0.10. When the fluid velocity change in the last 1,000 simulation steps was less than 2% for all cases, the simulations were assumed to have converged at that time. The time-averaged velocity value in the last 3,000 steps was calculated for further investigation.

To remove the complications arising from wettability effects, we assumed that the solid was completely nonwetting ($\theta = 180^{\circ}$) to the nonwetting fluid in all conditions. Given that condition, the wetting fluid coats the surface of the rock, whereas the nonwetting fluid is not in contact with the pore wall and can only occupy the central parts of the pores.

We applied a steady-state simulation condition, which assumes that the wetting fluid 218 saturation (S_w) and nonwetting fluid saturation (S_{nw}) are kept constant. In the initial state, both 219 fluids were specified as being randomly distributed in the pore spaces. The initial condition was 220 chosen because it allows each pore body to have roughly the same proportions of nonwetting and 221 wetting fluid according to the prescribed saturation; thus, this condition produces an extremely 222 uniform distribution of nonwetting and wetting fluid throughout the rock. In addition, because 223 224 the initial fluid connectivity is reduced under this initial condition and increases capillary 225 trapping, this initial distribution might be desirable in a CCS site (Fei Jiang & Tsuji, 2016). Under this initial condition, fluids are generated randomly in pore space; therefore, some 226 227 nonwetting fluids touch the grain surface at the initial stage. As the simulation progresses, the 228 wetting fluid coats the surface of the rock, and the nonwetting fluid moves to occupy only the central parts of pores because of the perfect wettability condition of the solid. A constant body 229

force was applied in the *z* direction to both fluids to mimic the pressure gradient: g = dP/dz (Fig. 1a). For the interaction between rock solid nodes and fluid voxels, no-slip boundary conditions were applied by using the halfway bounce-back scheme (X. Li et al., 2016; Singh et al., 2017). The density of both fluids was set at 1.0 in lattice units, which corresponds to 1,000 kg/m³ in the physical unit. We did this because our study was focused on viscosity differences, and the effect of density contrast is minor if inertial force can be neglected.

We applied a periodic boundary condition in the *x*, *y*, and *z* directions of our 3D model. Specifically, in the flow direction (*z* direction), the rock was mirrored to ensure that the pore spaces on the right side were connected to the left side of the digital rock (Fig. 1b) (F Jiang & Tsuji, 2017).

240 2.3.1 Relative permeability curve

The relative permeability curve is a plot of k versus saturation S_{nw} . As S_{nw} increases, k_{nw} increases and k_w decreases. The shape of this curve can change as a result of viscous drag force and capillary force; thus, an investigation of its variation with changes in M and Ca is important to determine the optimum conditions to achieve the desired value of k. We plotted k_{nw} and k_w as a function of S_{nw} to create relative permeability curves for various conditions.

In this study, we plotted all permeability curves for S_{nw} values within a wide range from 10% to 90%. Although some of these S_{nw} values might be unrealistic in actual injection settings because of irreducible wetting phase saturation (Tsuji et al., 2016), we still considered those hypothetical S_{nw} conditions for the ideal situation of a steady-state laboratory experiment.

250 2.3.2 M–Ca– k_{nw} permeability color map

251	After confirming the influence of <i>M</i> and <i>Ca</i> on k_{nw} and k_w in a two-phase flow system,
252	we conducted simulations at various M and Ca conditions with S_{nw} held at a constant value of
253	20% to create a plot of M – Ca – k_{nw} correlation or color diagram. The 20% S_{nw} condition was
254	chosen because it can realistically be achieved under all <i>M</i> – <i>Ca</i> conditions (Tsuji et al., 2016).
255	The color diagram is useful to evaluate the degree of influence of M and Ca on changes of
256	relative permeability when both parameters influence a system, and can also provide estimates of
257	relative permeability in a reservoir under a wide range of <i>M</i> and <i>Ca</i> .
258	3 Results and interpretation
259	Before conducting simulations in various M and Ca conditions, we first ran single-phase
260	simulations to determine the absolute permeability value of the porous media and to verify the
261	stability of the simulation. The results are presented in Appendix A. Afterwards, the effects of M
262	and Ca were then evaluated individually to see how they influence nonwetting and wetting
263	relative permeability curves in a two-phase flow system. Next, we conducted simulations over a
264	wide range of <i>M</i> and <i>Ca</i> values at a S_{nw} value of 20% to create a relative permeability color

266 3.1 Relative permeability curve from a two-phase simulation



Fig. 2. Relative permeability curves as a function of M at constant log $Ca = -0.25 \pm 0.1$ for M = 0.1-5: (a) nonwetting fluid relative permeability (k_{nw}) curve and (b) wetting fluid relative permeability (k_{w}) curve.

268 3.1.1 Effect of changes in M on k_{nw} and k_w

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We plotted values of k_{nw} for five values of M (0.1, 0.5, 1, 2, and 5) over the range of S_{nw} 269 (10% to 90%) while holding log Ca nearly constant at -0.25 ± 0.10 (Fig. 2a). We evaluated the 270 two-phase flow at high *Ca* to completely remove the capillary force effect. In all simulation 271 conditions, the Reynolds number must not exceed 10 (Bear, 1975). The maximum Reynolds 272 number reached in this set of simulations was 5.27 and 9.50 for nonwetting and wetting fluid, 273 respectively. The *M* values (0.1, 0.5, 1, 2, and 5) were chosen to represent a wide range of 274 mobility conditions, from an extremely unfavorable to an extremely favorable mobility 275 condition. The k_{nw} value increased as M increased, especially for intermediate S_{nw} values (30% to 276 70%). For M = 5, k_{nw} exceeded 1 under some S_{nw} conditions; that is, it exceeded the intrinsic 277 permeability of the rock. This result agrees with previous studies and can be explained by the 278

- lubrication effect, also known as the viscous coupling effect (Goel et al., 2016; Fei Jiang et al.,
- 280 2021; H. Li et al., 2005; Ramstad et al., 2010; Yiotis et al., 2007; Zhao et al., 2017).





281

282	To illustrate the lubrication effect, we provide a 2D image showing the distribution of
283	nonwetting fluid, wetting fluid, and solid rock at the final state of the simulation under various M
284	and S_{nw} conditions (Fig. 3). Because the wetting fluid (green) layer is generally present between
285	the nonwetting fluid and the rock, the wetting fluid moves along the rock surface and the
286	nonwetting phase is confined to the central part of the pore. Thus, the velocity of the nonwetting

fluid is affected only by the momentum transfer across fluid–fluid interfaces and not by contactwith the grain surface.

At sufficiently high values of M, the wetting fluid acts as a lubricant that enhances the 289 290 movement of the nonwetting fluid, making its permeability higher than in the single-phase condition such that k_{nw} is greater than 1 (Fig. 2a). This lubrication effect most strongly affects 291 k_{nw} in the intermediate S_{nw} range, when the wetting fluid forms thick films that provide the 292 293 nonwetting fluid with a moving boundary (Vafai, 2000). At low S_{nw} , the nonwetting phase tends to form droplets with low connectivity; thus, the lubrication effect is less pronounced. At high 294 S_{nw} , k_{nw} also decreases toward unity because less wetting fluid is present in the rock and the 295 296 nonwetting phase comes into contact with the rock, degrading the lubrication effect and reducing k_{nw} near to the single-phase condition. Because the lubrication effect occurs at high M values, it 297 is commonly observed in mixtures of heavy oil and water in EOR systems and has been 298 299 confirmed by experimental results (Goel et al., 2016; Shad et al., 2008).

In addition to the lubrication effect, the relative permeability change due to the viscosity 300 ratio can be linked to local pore-filling behavior. Because of the body force, a nonwetting fluid 301 droplet can move from one pore body to neighboring pore bodies. As M increases, as a result of 302 favorable mobility conditions, there is more chance for nonwetting fluid droplets to move to 303 several neighboring pore bodies through the narrow pore throats (Bakhshian et al., 2021), 304 305 causing fluid droplet fragmentation and thereby enabling the fluid to occupy smaller pore spaces. This might be one of the contributing factors causing k_{nw} to exceed 1 when M = 5 at intermediate 306 saturation. Furthermore, under high S_{nw} , nonwetting fluid already exists in most of the pore 307 308 bodies throughout the simulation time; thus, locally occurring pore-filling behavior effects decrease, causing k_{nw} to decrease towards unity under high S_{nw} conditions. 309



Fig. 4. Evolution of the number of clusters of nonwetting fluid at M = 1 and M = 5, with $S_{nw} = 20\%$ and log $Ca = -0.25 \pm 0.1$.

310

To investigate nonwetting fluid droplet fragmentation throughout the simulation, we 311 considered the evolution of the number of nonwetting fluid clusters during simulations with M =312 313 1 and M = 5, log $Ca = -0.25 \pm 0.10$, and $S_{nw} = 20\%$ (Fig. 4). Throughout the simulations, the number of nonwetting clusters for both simulation conditions decreases, suggesting that some of 314 the nonwetting fluid clusters become more connected and form larger clusters as the simulation 315 proceeds. In the final converged condition, the number of nonwetting fluid clusters is higher for 316 M = 5 than for M = 1. This result indicates that the nonwetting fluid at M = 5 forms more 317 318 individual clusters and has less fluid connectivity than at M = 1. This difference is the result of 319 the instability of the interface of the fluids due to the viscosity stratification (Yiantsios & Higgins, 1988; Yih, 1967). When two immiscible fluids have different viscosities, their 320 321 velocities will also be different at their interface, causing instability. The instability causes the nonwetting fluid to create more clusters. This result agrees with past research and our hypothesis 322 that instability at the fluid interface can cause fluid droplet fragmentation (Bischofberger et al., 323

324 2015; Mu'min et al., 2021; Nekouei & Vanapalli, 2017; Stone, 1994). As more fluid clusters are formed, the size of the nonwetting fluid clusters is smaller, and it is easier for them to pass 325 through the pore space. At M = 1, the nonwetting fluid is more connected and its fluid clusters 326 are larger. The larger clusters cannot pass through the small pore throats, leading to lower k_{nw} . 327

(a) M=1

328



Fig. 5. Two-dimensional slice showing the distributions of nonwetting (red), wetting fluid (green), and solid rock (black) at (a) M = 1 and (b) M = 5 at 20% S_{nw}. These images show a portion of the slice illustrated in Fig. 3.

329	To obtain further insights into fluid cluster size, we provide an illustration showing
330	nonwetting and wetting fluid distributions in an enlarged section of a 2D slice of the simulated
331	specimen for $M = 1$ and $M = 5$ at log $Ca = -0.25 \pm 0.10$ and 20% saturation after the simulation
332	had converged (Fig. 5). The nonwetting fluid (red) at $M = 1$ forms larger clusters in the pore
333	throat, whereas at $M = 5$, the nonwetting fluid forms a larger number of clusters but the size of
334	each cluster is smaller. This result is comparable to our findings (shown in Fig. 4) that
335	nonwetting fluid droplet fragmentation occurs as a result of instability at the fluid interface as M
336	increases. Because the nonwetting fluid clusters are smaller, it is easier for them to pass through

337	the pore space, leading to higher k_{nw} . Furthermore, the wetting fluid (green) coats the grain
338	surface under both conditions and produces a lubrication effect (Fig. 5).

Our results also demonstrate that k_w decreases as *M* increases (Fig. 2b), mainly when nonwetting fluid saturation is low. This phenomenon can be attributed to an increase of shear force from the nonwetting phase. As *M* increases and nonwetting fluid droplet fragmentation occurs, the wetting fluid has a larger contact area with the nonwetting fluid (Fig. 5). As a result, the wetting fluid receives a higher shear force from the nonwetting fluid, which inhibits the flow of the wetting fluid so that it has a lower velocity. In addition, as wetting fluid saturation decreases, the k_w gaps under all *M* conditions become smaller.

To confirm our interpretation, we calculated the degree of contact between nonwetting 346 and wetting fluid by calculating the number of wetting fluid voxels that were adjacent to a 347 nonwetting fluid voxel, for conditions of M = 1 and M = 5 at log $Ca = -0.25 \pm 0.10$ and $S_{nw} =$ 348 20%. When the simulations converged, 13.77679% and 13.88825% of wetting fluid voxels were 349 adjacent to nonwetting fluid voxels at M = 1 and M = 5, respectively. This result shows that, at M 350 = 5, the wetting phase fluid has a higher degree of contact with the nonwetting fluid and receives 351 more drag force from the nonwetting fluid; thus, k_w is lower at M = 5 than at M = 1, as shown in 352 Fig. 2b. 353

In addition, Zhao et al. (2017) suggested that the k_w decrease in response to an *M* increase occurs because the wetting fluid is more attached to the grain surface when the viscosity ratio is large. When the viscosity ratio is high, fragmentation of the nonwetting phase tends to occur easily and the fragments move towards the centers of pore bodies, so that wetting fluid can easily move to coat the grain surface. Consequently, because the wetting fluid has a larger contact area with the grain surface, it receives more shear drag force, which causes a decrease in velocity. To

360 confirm our interpretation, we calculated the degree of contact between the wetting fluid and the 361 grain surface, as indicated by the number of wetting fluid voxels having a solid surface as a 362 neighbor, for M = 1 and M = 5 at log $Ca = -0.25 \pm 0.10$ and $S_{nw} = 20\%-70\%$ after the simulation 363 had converged (Table 1).

	<i>M</i> =1	<i>M</i> =5
Snw=20%	36.79546%	36.74534%
Snw=50%	50.94340%	51.18913%
Snw=70%	67.81628%	68.25865%

Table 1. Percentage of wetting fluid that has a grain surface as a neighbor voxel fluid at M = 1 and M = 5, with S_{nw} of 20%–70% and log $Ca = -0.25 \pm 0.1$, after the simulation has converged.

364

At $S_{nw} = 20\%$, the percentage of wetting fluid voxels adjacent to a solid surface is low, 365 because the wetting fluid film between the nonwetting fluid and grain surface is thick (Fig. 3) 366 367 and only a small portion of the wetting fluid is in direct contact with the grain surface. The values for M = 1 and M = 5 are also similar, suggesting that the nonwetting fluid at low S_{nw} has 368 less chance to approach the rock surface because of the thick wetting fluid film even as M 369 370 increases. Thus, at low S_{nw} , the dominant cause of the k_w decrease is the shear force at the fluid interface and not the shear drag force at the wall. When $S_{nw} = 50\%$ and 70% and M = 5, the 371 proportion of the wetting phase fluid attached to the grain surface increases compared to when M 372 = 1, indicating that the wetting fluid has a higher degree of contact and receives more drag force 373 from the solid wall. Thus, k_w at M = 5 is lower than that at M = 1 (Fig. 2b). 374



Fig. 6. Normalized velocity fields for the nonwetting fluid in a two-phase simulation with (a) M = 1 and (b) M = 5, at log $Ca = -0.25 \pm 0.1$ and $S_{nw} = 20\%$.

375

The results of fluid velocity for *M*=1 and *M*=5 in the pore spaces at 20% saturation after the simulation converges are plotted in Figs. 6a and 6b, respectively. The fluid velocity field in Fig. 6 is normalized to consider the effect of viscosity change using the equation $U^* = \frac{U}{\Delta P L/\mu}$. It 379 can be seen that the normalized fluid velocity is higher at M = 5 than at M = 1, indicating that k_{nw}





Fig. 7. Relative permeability curves for the nonwetting (k_{nw} , green) and wetting fluids (k_w , blue) for log Ca = -0.25 to -3.5 with M = 1.

381

382 3.1.2 Effect of *Ca* change on k_{nw} and k_w

The k_{nw} and k_w curves for six different values of log *Ca* (-0.25, -1.25, -2.25, -2.65, -384 3.00, and -3.50 ± 0.10) at *M* = 1 are plotted (Fig. 7). The *Ca* value is altered by changing the IFT value. It was difficult to keep *Ca* constant because *Ca* is also a function of fluid velocity, which is a direct result of the simulation. Thus, we allowed a small error of ± 0.10 for *Ca* under all conditions.

The k_{nw} values remain relatively constant between log Ca = -0.25 and -1.25, i.e., for high *Ca*/low IFT values, and the relative permeability curve for the nonwetting fluid is an approximately straight line, in agreement with previous studies (Asar & Handy, 1989; Shen et

391	al., 2010). However, as log <i>Ca</i> becomes lower than -1.25 , k_{nw} notably decreases as <i>Ca</i> decreases.
392	This decrease occurs because the flow of the nonwetting fluid is more strongly inhibited by
393	capillary force when Ca is low. Capillary force is controlled by IFT, the surface tension forces
394	between the nonwetting and wetting fluids. As IFT increases, the nonwetting fluid becomes
395	trapped by the larger capillary force in the small pore spaces, thus causing k_{nw} to decrease. This
396	result also suggests that k_{nw} does not decrease linearly with changes in IFT/ <i>Ca</i> , because the k_{nw}
397	decrease gap gradually becomes larger as Ca decreases. This phenomenon occurs because as Ca
398	becomes lower, the influence of capillary force becomes more dominant.
399	For the wetting fluid, k_w also decreases as <i>Ca</i> decreases (Fig. 7), but at a much smaller
400	rate compared to k_{nw} , consistent with previous research (Harbert, 1983; Fei Jiang et al., 2014;
401	McDougall et al., 2007; Ramstad et al., 2010; Zhao et al., 2017). The reason for this result is that
402	the wetting fluid flows only along the rock surface and the nonwetting fluid flows in the central
403	part of the pores (Fig. 3). Thus, the wetting fluid is mainly affected by interaction with the rock
404	surface, and the change in capillary force has a negligible effect. However, when Ca is low (log
405	$Ca = -3.50$), the k_w decrease is more notable. This phenomenon can be attributed to the drag

407 to other *Ca* conditions and the nonwetting fluid velocity is low. This result suggests that the

408 influence of the nonwetting fluid drag force on the wetting fluid increases, causing k_w to 409 markedly decrease with the k_{nw} decrease. Nevertheless, we conclude that the variation in k_w is

410 dominated by the fluid saturation in the system and is little affected by changes in IFT or *Ca*.

411 $3.2 M - Ca - k_{nw}$ color diagram

412	After confirming the influence of <i>M</i> and <i>Ca</i> on k_{nw} and k_w in a two-phase flow system,
413	we conducted simulations with various values of M and Ca at a constant 20% S_{nw} to create a M -
414	<i>Ca</i> - k_{nw} correlation diagram (Fig. 8). We chose the 20% S_{nw} condition because it is realistic under
415	all <i>M</i> – <i>Ca</i> conditions (Tsuji et al., 2016). We created the color diagram only for the nonwetting
416	fluid because the relative permeability curves (Fig. 7) demonstrate that k_w does not respond
417	markedly to changes in Ca. We also created a regression model and equation for our results
418	reported in this section (see Appendix B). The maximum Reynolds number reached in this set of
419	simulations was 3.92 for $M = 0.10$ and log $Ca \approx 0$.
420	Color maps are useful to provide estimates of k_{nw} under a wide range of M and Ca
421	conditions in a reservoir and to analyze the degree of influence on k_{nw} by M and Ca (Fig. 8). The
422	first color map (Fig. 8a) depicts the parameter space of $\log M = -1.02$ to 0.70 and $\log Ca = -4.00$
423	to 0.00, which represents the broad range of conditions that can be simulated. Values higher than
424	this range are hard to achieve at the field scale, and lower values are outside the stable range of
425	the simulation. The second color map (Fig. 8b), covering the lower left quadrant of the parameter
426	range shown in Fig. 8a with parameter space of $\log M = -1.02$ to 0.00 and $\log Ca = -4.00$ to -
427	1.00, was produced to increase the accuracy of k_{nw} estimates for a system with low M and low
428	Ca, which is common in CCS field.

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Fig. 8. Relative permeability map of the nonwetting fluid for (a) $\log M = -1.02$ to 0.70 and $\log Ca = -4.00$ to 0.00, and (b) $\log M = -1.02$ to 0.00 and $\log Ca = -4.00$ to -1.00. The dots indicate the *Ca*-*M* conditions used for LBM simulation.

429

430 In the specified parameter range, k_{nw} varies from 0.0002 to 0.6125 (Fig. 8a). For high log 431 Ca values (between -1.25 to 0.00), k_{nw} does not vary with log Ca and is only influenced by M, as indicated by the almost straight vertical borders between color bands. This finding agrees with 432 our results from section 3.1.2 (Fig. 7), which shows that for log $Ca \ge -1.25$, the nonwetting fluid 433 relative permeability curve is not affected by Ca. However, as log Ca becomes lower than -1.25434 (*Ca* value ≈ 0.06), k_{nw} can be seen to vary on both the x axis (log *M*) and the y axis (log *Ca*), that 435 is, k_{nw} starts to be affected by the capillary force. The diagram agrees with our finding in section 436 3.2 that for log Ca lower than -1.25, k_{nw} decreases as Ca decreases. 437

438	The k_{nw} variation with M (x axis) is more pronounced for log $M > 0$ ($M > 1$) than for log
439	M < 0 (Fig. 8a). This result is consistent with our findings in section 3.1 and is caused by the
440	lubrication effect resulting from viscosity differences. When μ_{nw} is higher than μ_{w} , the resulting
441	instability and viscous coupling effect enhance the relative permeability of the nonwetting fluid.
442	The diagram clearly captures the contrast between k_{nw} at log $M < 0$ and at log $M > 0$.
443	To check the detailed k_{nw} variation under low <i>M</i> and low <i>Ca</i> conditions, in Fig. 8b we
444	show an enlargement of part of the lower left quadrant of the parameter range illustrated in Fig.
445	8a. The k_{nw} value range is 0.0002 to 0.2725 in Fig. 8b, and in this parameter space, both <i>Ca</i> and
446	<i>M</i> influence k_{nw} and thus the borders between the color bands are curved. In a CCS field, <i>M</i> is
447	usually lower than 1 and Ca is generally low (Zheng et al., 2017). Therefore, Fig. 8b is useful for
448	estimating nonwetting fluid relative permeability in a CCS field.
449	The diagram is useful for evaluating k_{nw} when the effects of M and Ca are considered
450	simultaneously, and the graph neatly summarizes all the main results of this study. The diagram
451	shows that capillary force starts affecting the flow of the nonwetting fluid when log Ca is less
452	than -1.25 and that the effect of changes in <i>Ca</i> may be just as important as that of changes in <i>M</i> .
453	The graph also shows that k_{nw} is affected by <i>M</i> under all conditions, but most markedly when log
454	$M > 0$. The graph yields a good estimate of k_{nw} at 20% saturation based on the M and Ca
455	parameters.

456 **4 Discussion**

We have demonstrated how *M* and *Ca* can markedly change the relative permeabilities of
the nonwetting and wetting fluids in a reservoir; thus, our results can help identify the optimum
properties of the immiscible fluids to be used in a geologic reservoir. For example, in a CCS

460	project, the injectivity of the CO ₂ is highest when the reservoir fluid has a lower viscosity and the
461	IFT between the two fluids is low. Thus, the relative permeability of injected CO ₂ is higher in a
462	saline aquifer than in an oil field. For an EOR project, the relative permeability of oil is higher
463	when a fluid with low viscosity is injected.

The relative permeability map (Fig. 8) is useful to provide accurate estimates of k_{nw} in 464 reservoir-scale simulations. Currently, the relative permeabilities of the nonwetting and wetting 465 466 fluid are simulated on the basis of a uniform relative permeability curve, without regard to the M and *Ca* conditions. However, we have shown that the nonwetting fluid, at a typical saturation 467 (S_{nw}) of 20%, can vary in relative permeability by an order of magnitude, from 0.0002 to 0.6125, 468 depending on M and Ca conditions. The color map created in this study can provide more 469 accurate estimates of relative permeability (e.g., temporal permeability variations in a 3D 470 reservoir model) if Ca and M are derived from the reservoir simulation. In addition, although the 471 472 M generally remains constant in the two-phase flow, the Ca value can greatly changes depending on the distance from the injection well in a reservoir. For example, when evaluating the reservoir 473 474 area located near the injection well, the injection pressure creates a high-pressure gradient which causes the injected fluid velocity to be high. As the fluid flows away from the injection site, the 475 fluid velocity becomes slower, thus the Ca becomes lower. This means that fluid relative 476 477 permeability varies based on location inside the reservoir, and the permeability variation can be evaluated using the relative permeability map, thus providing a more accurate relative 478 permeability estimation. 479

In this study, we conducted the simulation under steady-state conditions, as being most representative for illustrating fluid flows in a reservoir area distant from the injection site, where boundary effects are negligible (Honarpour et al., 2018; Ramstad et al., 2012). In addition, under

steady-state conditions, relative permeability can be measured directly by using Darcy's law; 483 thus, the relative permeability value is more accurate than one calculated under non-steady-state 484 485 conditions. However, there are some differences between relative permeability values obtained under steady-state and non-steady-state conditions, and there are also some limitations to steady-486 state simulations. Under steady-state conditions, both fluid phases coexist together in the 487 488 reservoir at the prescribed saturation from the beginning of the simulation. In contrast, under non-steady-state conditions, the reservoir is initially filled with wetting fluid, and nonwetting 489 fluid is injected and gradually displaces the wetting fluid, which results in a gradual increase of 490 nonwetting fluid saturation and a decrease of wetting fluid saturation. As a result, under non-491 steady-state conditions several factors can affect relative permeability variation, such as 492 fingering phenomena, drainage and imbibition conditions, and the pore-filling behavior during 493 the injection (Bakhshian et al., 2021; Rabbani et al., 2017). Therefore, an unsteady-state 494 simulation can better represent relative permeability near the borehole or injection site, where 495 remarkable displacement occurs. Nevertheless, if considering the law of two-phase fluid flow 496 that coexist and flow together throughout the whole drainage area, steady-state relative 497 permeability curves are more suitable for planning the whole reservoir design. In addition, 498 relative permeability obtained under steady-state conditions is more accurate because the relative 499 permeability can be directly measured by using Darcy's law. In contrast, in an unsteady-state 500 simulation, relative permeability must be calculated from the capillary pressure change and 501 production data under several assumptions (e.g., the Johnson, Bossler, and Naumann method; 502 (Esmaeili et al., 2020; Johnson et al., 1959)). As a result, the obtained relative permeability is 503 504 less accurate. Steady-state condition is also better to estimate relative permeability for CCS

505 monitoring system after injection, as at the time, the CO₂ will coexist with the wetting fluid in 506 the reservoir.

In this study, we created a relative permeability map for $S_{nw} = 20\%$, which is a reasonable 507 508 condition that can be achieved in all or most systems. As S_{nw} increases, k_{nw} also increases until it reaches its maximum value (e.g., k_{nw} at the irreducible saturation). Therefore, it is advisable to 509 create relative permeability maps for several saturation conditions. One possible future direction 510 511 from this study would be to create a four-dimensional $M-Ca-S_{nw}-k_{nw}$ graph to yield k_{nw} estimates for all saturation conditions. Nevertheless, the great consistency of variations in k 512 caused by changes in M and Ca found in this study suggests that maps for other saturation 513 conditions will have similar features to Fig. 8. It would also be possible to create a regression 514 model for $k_{nw} = f(M - Ca - S_{nw})$ to estimate k_{nw} under all saturation conditions. 515

In this study, we produced a relative permeability diagram for a digital specimen of Berea 516 sandstone. In addition to M and Ca, the influences upon relative permeability include the pore 517 geometry of the rock, such as pore size distribution, pore connectivity, and other parameters 518 (Jiang et al. 2018, WRR). These parameters differ among rock formations, meaning that the 519 520 relative permeability maps of different types of reservoir rocks will vary. Our methodology makes it possible to create accurate maps of relative permeability for other reservoir rocks. 521 In this study, we used log M ranging from -1.02 to 0.70 and log Ca ranging from -4.00522 to 0.00; however, in the CO₂ injection process, log M might reach as low as -1.8 and log Ca 523 524 might reach -6.8 (Zheng et al., 2017). Such values are outside the stable range where this 525 simulation can be applied. Nevertheless, the objective of this study was just to propose a method to predict the relative permeability of a two-phase flow fluid in a reservoir by using a color map 526 527 diagram to evaluate the effect of the viscosity ratio and capillary number (Fig. 8). In a future

study, we plan to improve the stability range of the simulation to allow lower *M* and *Ca* values tobe considered.

In geological CO_2 storage, the maximum amount of CO_2 that can be stored and the 530 injectivity of the CO_2 into the reservoir must both be considered (Tsuji et al., 2016). Thus, the 531 results of this study must be combined with information on the effects of M and Ca on the 532 maximum saturation of the nonwetting fluid. In this study, we showed that, for CO₂ as the 533 534 nonwetting fluid, the relative permeability increases as M increases and decreases as Ca becomes very small. In contrast, Tsuji et al. (2016) demonstrated that the maximum S_{nw} increases as M 535 increases and notably increases at low Ca. Thus, although a high value of M is desirable to 536 increase both the CO₂ capacity and injectivity, a low *Ca* value can also increase the maximum 537 CO₂ saturation but at the cost of reduced relative permeability. Both factors must be taken into 538 account when choosing suitable conditions for CO₂ storage. 539

An advantage of using M and Ca is that both parameters are dimensionless, meaning that 540 the results obtained in this pore-scale study can potentially be upscaled to the reservoir scale. 541 Ideally, the pore-scale results are also valid at the reservoir scale as long as the ratios of the 542 parameters (viscosity and IFT) are maintained for both fluids, because the fluid flow behavior at 543 the reservoir scale is controlled by the fluid dynamics at the pore scale. However, this ideal is 544 challenged by the inhomogeneity of the porous medium. The relative permeability is likely to 545 546 vary throughout the reservoir due to differences in pore size, pore connectivity, and many other factors. Nevertheless, the results of a pore-scale simulation are important to verify relative 547 permeability variations arising from selected factors (in this study, M and Ca) by eliminating 548 549 other factors. The results of a pore-scale study of relative permeability could then be upscaled by considering the structural factors of the reservoir, e.g., its porosity and pore connectivity, usingadvanced techniques such as machine learning.

552 5 Summary

553	To evaluate the influence of viscosity ratio M and capillary number Ca on relative
554	permeability k in a two-phase flow system, we calculated k for nonwetting and wetting fluids
555	$(k_{nw} \text{ and } k_w)$ under various <i>M</i> and <i>Ca</i> conditions using an LBM simulation. The main results of
556	this study are as follows.

- 1) In our simulations, the relative permeability of the nonwetting fluid increased as the
- viscosity ratio increased due to the lubricating effect, locally occurred pore-filling
- behavior, and instability of the fluid interface. Specifically, at high viscosity ratios (M =

560 5), k_{nw} could exceed 1 as a result of the lubricating effect.

- 561 2) The relative permeability of the wetting fluid decreased as *M* increased due to the562 increase in shear force from the nonwetting fluid (viscous coupling effect)
- 3) As *M* increased, the number nonwetting fluid clusters became higher, indicating that the
 nonwetting fluid became more disconnected and the size of each clusters becomes
 smaller.
- 566 4) At high capillary numbers (log Ca = -0.25 to -1.25), k_{nw} did not respond to Ca, however 567 as log Ca becomes lower than -1.25, k_{nw} decreases as Ca decreases due to the capillary 568 force effect.
- 569 5) As *Ca* decreased k_w decreased, but at a negligible rate compared to k_{nw} .

570 6) k_{nw} can change markedly in a wide range of *M*–*Ca* parameter space, and the *M*–*Ca*– k_{nw} 571 correlation map created in this study can provide k_{nw} estimates at various reservoir 572 conditions.

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582 Appendices

583 A. Single-phase simulation

Before conducting the two-phase flow simulations, we first ran single-phase simulations to calculate the absolute permeability of the 3D rock model. Single-phase simulations can also be used to verify the accuracy and precision of the simulation. When only one type of fluid exists in a rock, the absolute permeability can be calculated from Darcy's law:

588
$$k = \frac{v \,\mu \,\Delta l}{\Delta P} \qquad (A.1)$$

where *k* is the absolute permeability (m²), *v* is the average fluid velocity (m/s), μ is the dynamic viscosity of the fluid (Pa s), Δl is the distance between the inlet and the outlet, and ΔP is the applied pressure difference between the inlet and the outlet. In this study, because the body force was applied only in the *z* direction, we calculated *k* in the *z* direction (k_z).

593 Because all simulation units are in lattice Boltzmann units, they must subsequently be 594 converted into physical units. The unit conversion for k can be performed using the formula:

595
$$k_{(physical)} = k_{(LB)} * A^2$$
 (A.2)

596 where *A* is the resolution of the digital rock, i.e. the physical size of a lattice grid (2.673 μ m in 597 this study).

Absolute permeability is an intrinsic property of a rock that is independent of the type of fluid. The single-phase simulations were conducted with several different values of fluid viscosity and body force to ensure the consistency of the results. When the fluid summation of velocity change in 1,000 steps difference was less than 2% for all cases, the simulations were assumed to have converged at that time. The absolute permeability results in various viscosity and body force conditions are shown in the table A.1.

kinematic viscosity	Body force	absolute permeability
(lattice-Boltzmann unit)	(lattice-Boltzmann unit)	(Darcy)
0.155555	0.0001	1.3029
0.155555	0.0002	1.3025
0.310000	0.0001	1.3042
0.310000	0.0002	1.3038

604

Table A.1 single-phase simulation conditions and results

The absolute permeability of the Berea rock is 1.3034 ±0.001 Darcy. The absolute
permeability value remains constant under various conditions, which demonstrates the accuracy
and stability of the simulation. The result obtained from this simulation is slightly different from

the result by the Dong and Blunt (2009), which gives the absolute permeability value of 1.193 608 Darcy. The difference might be due to the difference in size of digital rocks. Dong and Blunt 609 used digital rock with dimension of 400x400x400 voxels with resolution of 5.345 µm (9.772 610 mm³ in actual size), while we used digital rock with dimension of 400x400x400 voxels with 611 resolution of 2.673 µm (1.222 mm³ in actual size) is used. The dimension difference might have 612 613 caused a slight change in the rock sample heterogeneity. In addition, Dong and Blunt applied Pore Network Model (PNM) to calculate the absolute permeability, while in this study, LBM 614 simulation is used. Nonetheless, the absolute permeability value obtained is still in the same 615 616 order as described by the paper.



Fig. A. Velocity fields for single phase simulation at (a) μ = 0.155555 and (b) μ = 0.310000 and normalized velocity fields for single phase simulation at (c) μ = 0.155555 and (d) μ = 0.310000.

617

We show the velocity field of single-phase simulation for kinematic viscosity = 0.155555618 and 0.310000 with equal body force of 0.0001 (Fig. A(a) and A(b)). The fluid velocity for 619 simulation with fluid kinematic viscosity of 0.155555 nu {LB} is significantly higher compared 620 to the fluid kinematic viscosity of 0.310000 nu {LB} case. This is because fluid average 621 velocity is inversely proportional to viscosity. Thus, fluid velocity is not proportional to 622 623 permeability, and must be normalized into dimensionless velocity field. Fig. A(c) and A(d) show the normalized fluid velocity field for both conditions. The normalization is done using the 624 equation $U^* = \frac{U}{\Delta PL/\mu}$. Both figures show a relatively similar results, which indicate that a similar 625 626 absolute permeability value is obtained from both conditions. 627

B. Regression model of the M-Ca- k_{nw} relationship 628

We created a regression model to generate an empirical equation that can be used to 629 calculate k_{nw} as a function of M and log Ca at 20% S_{nw}. A height map of our simulation results 630 and the fitted polynomial regression curve is shown in Fig. B. 631



Fig. B. Nonwetting fluid relative permeability height map (dots) and the fitted polynomial regression curve.

Past research has shown that k_{nw} can be estimated as a function of M by using an equation with second-degree polynomial (equation 3 and 4 (Goldsmith & Mason, 1963)); thus, we chose a second polynomial equation for the regression towards the M value. For the log *Ca* value, we also set it as second-degree polynomial to prevent overfitting.

637 Based on our model, k_{nw} can be calculated with the following formula:

638

639
$$k_{nw} = 0.05983 + 0.1844 M - 0.08714 (log Ca) - 0.009922 M2 + 0.0275M (log Ca)
640 - 0.03335 (log Ca)2$$

641

- 642 Our model has an *R*-squared value of 0.9481 and a summed square of residuals value of643 0.09871.
- 644 Because we created a color map diagram for only one saturation condition ($S_{nw} = 20\%$),
- 645 we only created a regression model to calculate k_{nw} based on *M* and *Ca* values for this saturation
- 646 condition. Because the relative permeability increase with respect to saturation is not linear, we
- 647 expect the regression model to be different for other saturation conditions. In the future, we plan
- to create more relative permeability color maps for various saturation conditions and create a
- 649 four-dimensional log M-log Ca- S_{nw} - k_{nw} graph to yield k_{nw} estimates for all saturation
- 650 conditions. Then, we will attempt to create a regression model to estimate k_{nw} as a function of M,
- 651 log Ca, and S_{nw} .
- 652

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858	Figure Caption
859	Fig. 1. Berea sandstone digital rock model: (a) core form and (b) mirrored form.
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861 862	Fig. 2. Relative permeability curves as a function of <i>M</i> at constant log $Ca = -0.25 \pm 0.1$ for $M = 0.1-5$: (a) nonwetting fluid relative permeability (k_{nw}) curve and (b) wetting fluid relative permeability (k_w) curve.
863	
864 865	Fig. 3. Two-dimensional slice showing the distributions of nonwetting fluid (red), wetting fluid (green), and solid rock (black) at various values of M and S_{nw} .
866	
867 868	Fig. 4. Evolution of the number of clusters of nonwetting fluid at $M = 1$ and $M = 5$, with $S_{nw} = 20\%$ and log $Ca = -0.25 \pm 0.1$.

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870 871 872	Fig. 5. Two-dimensional slice showing the distributions of nonwetting (red), wetting fluid (green), and solid rock (black) at (a) $M = 1$ and (b) $M = 5$ at 20% S_{nw} . These images show a portion of the slice illustrated in Fig. 3.
873	
874 875	Fig. 6. Normalized velocity fields for the nonwetting fluid in a two-phase simulation with (a) $M = 1$ and (b) $M = 5$, at log $Ca = -0.25 \pm 0.1$ and $S_{nw} = 20\%$.
876	
877 878	Fig. 7. Relative permeability curves for the nonwetting (k_{nw} , green) and wetting fluids (k_w , blue) for log $Ca = -0.25$ to -3.5 with $M = 1$.
879	
880 881 882	Fig. 8. Relative permeability map of the nonwetting fluid for (a) $\log M = -1.02$ to 0.70 and $\log Ca = -4.00$ to 0.00, and (b) $\log M = -1.02$ to 0.00 and $\log Ca = -4.00$ to -1.00 . The dots indicate the <i>Ca</i> - <i>M</i> conditions used for LBM simulation.
883	
884 885	Fig. A. Velocity fields for single phase simulation at (a) $\mu = 0.155555$ and (b) $\mu = 0.310000$ and normalized velocity fields for single phase simulation at (c) $\mu = 0.155555$ and (d) $\mu = 0.310000$.
886	
887 888	Fig. B. Nonwetting fluid relative permeability height map (dots) and the fitted polynomial regression curve.
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