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### Key Points:

- Direct pore-scale simulation of spontaneous imbibition (SI) in a 3D pore space extracted from a tight reservoir rock
- SI under reservoir condition is about two times faster than that under ambient condition mainly due to a lower viscous force
- Snap-off of oil droplets during the SI mainly occurred in pore bodies surrounded by slit-shaped pore throats

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## Investigation of Spontaneous Imbibition Behavior in a 3D Pore Space Under Reservoir Condition by Lattice Boltzmann Method

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**Abstract** Spontaneous imbibition (SI) of the injected fluid into the pore space of a tight oil reservoir and replacing the crude oil therein has been considered as one of the possible mechanisms in increasing oil recovery. Such deeply buried reservoir rocks is usually under high-pressure and high-temperature conditions. Besides, their interior porous structures are usually characterized as irregular pore bodies and slit-shaped pore throats. As a result, an accurate description of the SI behavior driven by capillary force in the real pore space under reservoir conditions is crucial to understand the process and uncover the controlling mechanisms. An improved multi-component pseudo-potential lattice Boltzmann method was developed to simulate the SI behavior in a representative 3D pore space extracted from a tight sandstone reservoir rock. Comparison of the SI behavior under ambient condition and reservoir condition showed that the latter case exhibited two times faster of the imbibition. Moreover, a snap-off of the oil droplet phenomenon was observed in the pore bodies surrounded by slit-shaped pore throats. The snap-off oil droplets stuck in pore bodies accounted for 9.47% of the pore volume. These results indicated the importance of investigating the SI in a real porous structure and under actual reservoir condition. The proposed pore-scale simulation method provides a useful tool in understanding the complex SI pattern and the resulted enhanced oil recovery.

**Plain Language Summary** Tight oil reservoir has become an important world energy supply due to formation stimulations such as hydraulic fracturing. In the process, a large amount of water was injected and retained in the formation. It is believed that the capillary force induced spontaneous imbibition (SI) of the residual water into the tiny pore space is beneficial to the oil recovery in the tight reservoir. However, an accurate description of the process that happened deep underground is difficult and the controlling mechanism remains to be fully understood. Here, pore-scale simulations were performed to directly visualize the detailed SI behavior under the reservoir and ambient conditions. The results showed (1) faster SI appeared under the reservoir condition, (2) snap-off of the oil droplets occurred in large pores surrounded by slit-shaped pores. These findings suggest that the reservoir temperature and pressure and the detailed pore morphology have a huge impact on the SI behavior.

## 1. Introduction

Unconventional oil reservoirs, which are previously classified as uneconomic resources by traditional developing methods, have become one of the important world energy supplying parts in recent years with the application of horizontal drilling, hydraulic fracturing and numerous enhanced oil recovery (EOR) methods (K. Singh et al., 2019; Xie et al., 2021). These oil reservoirs usually bury deep underground with conditions characterized as high-pressure and high-temperature. In a typical unconventional oil reservoir exploitation, a hydraulic fracturing job is proceeded after the completion of horizontal drilling to increase transport channels for oil recovery (Z. Chen et al., 2020). Previously, a flowback operation of the fracturing fluid is usually designed to decrease the potential reservoir damage. However, recent developments in these oil fields showed that partly flowback or even no flowback with the subsequent shut-in operation increased the oil recovery (Abbasi et al., 2014; Bertonecello et al., 2014; J. Liu et al., 2019).

Spontaneous imbibition (SI) of the injected fluid into the reservoir pore space and displacement of the crude oil therein are claimed as one of the principal reasons for the increased oil recovery in the unconventional

reservoir during the shut-in operation (K. Singh et al., 2019). The SI is a particular type of multiphase flow driven by capillary force. For a typical tight rock, the characteristic radius of its pore space is very small, which ranges from tens nanometers to tens micrometers (Lai et al., 2018; Nelson, 2009). According to the Young and Laplace equation, capillary force becomes one of the dominant factors that control the flow in such a small-scale porous media. Based on whether the flow directions of imbibed-in wetting phase and driven-out non-wetting phase are the same or opposite, the SI is classified as co-current SI and counter-current SI. In a hydrophilic tight reservoir rock, the SI of the injected aqueous fluid would replace the crude oil in the pore space.

Several core scale laboratory studies have verified the potential of increasing oil recovery by SI (Abbasi et al., 2014; Bertoncello et al., 2014; Habibi et al., 2015; H. Singh, 2016). For instance, Lan et al. (2015) compared the SI in intact and crushed samples of tight rocks and argued that rock fabric and pore connectivity played a crucial role in the SI behavior. Nuclear magnetic resonance (NMR) measurement of the  $T_2$  spectra of tight reservoir rocks during the SI showed that distinct recovery performances were achieved in different size pores (Cheng et al., 2018; X. Wang et al., 2018). Hodder and Nychka (2019) investigated the influence of wettability on the SI behavior in 3D printed sandstone samples. Haugen et al. (2014) pointed out co-current SI was more important than counter-current SI for oil recovery. Moreover, adding surfactant or nanoparticles in the imbibed fluid was argued to be beneficial to the oil recovery (Dai et al., 2017; Mohammadi et al., 2019; Zhu & Li, 2020). For instance, Xu et al. (2019) investigated the contribution of interfacial tension (IFT) reduction and wettability alteration on EOR separately during the SI and pointed out that ultralow IFT and increasing the hydrophilicity of the sample were desirable for the EOR. Besides, the reservoir temperature and pressure have non-negligible influences on the in-situ fluid properties such as viscosity and IFT (Kalantari Meybodi et al., 2016; Strand et al., 2008; S. Yang, 2017; Zeppieri et al., 2001). An accurate description of the SI behavior was complicated due to the geometrical complexity of the porous structure and the in-situ high-pressure and high-temperature conditions. There is not a consensus on the relative importance of the influence factors based on the core scale experiments (Kibria et al., 2018; Kuang et al., 2020). A detailed investigation of the SI at the pore-scale under reservoir conditions is crucial in uncovering the controlling mechanisms of fluids imbibed-in and driven-out from the rock.

Several pore-scale theoretical models, experiments, and simulations have been conducted to investigate the SI behavior. Such investigations of the detailed pore-scale SI behavior under reservoir conditions are necessary to unveil the complex flow dynamics in the unconventional reservoir. The classical Lucas-Washburn (LW) equation, which can be dated back to the early twentieth century, stated that the SI length in a capillary tube is proportional to  $\sqrt{t}$  (Washburn, 1921). However, actual pore geometry in tight rocks is far from a collection of cylindrical tubes and the applicability of LW law can be questioned (Ponomarenko et al., 2011). Reyssat et al. (2008) demonstrated the SI behavior in channels with axial variations followed  $l \propto \sqrt{t}$  at short times, whereas followed  $l \propto \sqrt[4]{t}$  at longer times. They also argued SI rate in porous media will depend on the spatial variation of the permeability. Ponomarenko et al. (2011) indicated that the capillary rise in the corners of irregular pores followed  $l \propto \sqrt[3]{t}$ . Primkulov et al. (2020) stated that a large fraction of dissipation happened near the contact line and dynamic contact angle should be used for the accurate descriptions of the SI behavior. A recent review of LW law based modeling of the SI was presented by Cai et al. (2021). Although it was found that the SI in a homogeneous porous media generally obeyed the LW law, the pore space in an unconventional tight reservoir rock showed much more heterogeneity and geometrical irregularity, and whether the LW law still applicable needs to be further investigated (H. Singh, 2016; X. Wang et al., 2018; R. Yang et al., 2017).

Developments in the visualization of pore-scale multiphase flow behavior were facilitated by accurate fabrication of micromodels and fast development of imaging methods (Bazaikin et al., 2017; Cao et al., 2020; David et al., 2011; Lei et al., 2020; Y. Zhao et al., 2017). Visualization of the detailed SI processes in delicate 2D transparent models enabled several new discoveries of the transport mechanisms (Zhang et al., 2011; B. Zhao et al., 2016; Zheng et al., 2021). For instance, the SI in glass beads models showed that the pore size distribution played significant roles in the propagation pattern (Ashraf et al., 2017) and the residual oil distribution (Hatiboglu & Babadagli, 2008; Meng et al., 2016). The micromodel with structural details of the actual pore space in a rock was used to study the pore-level physics of the SI (Meng et al., 2016; Sun et al., 2016). It was found the pore geometry has a non-negligible influence on the imbibition morphology.

The fast development of 3D imaging methods, such as magnetic resonance imaging (MRI) and X-ray computer tomography (CT), enabled the direct observation of the SI inside reservoir rock samples (Armstrong et al., 2014; Bazaikin et al., 2017; David et al., 2011; Fernø et al., 2013). For instance, Fernø et al. (2013) visualized the details of SI front by MRI and pointed out that the core heterogeneities had a dominant effect on the SI behavior. Zhao et al. (2017) employed the CT and neutron radiography to characterize the influence of pore structure on the water imbibition in tight sandstones. Moreover, the 3D images of the flow evolution inside a rock sample have been reported by using high spatial and temporal resolution CT (Alhammadi et al., 2020; Alzahid et al., 2020; Gao et al., 2020; Iglauer & Lebedev, 2018; Joyce et al., 2015; Ju et al., 2018). Singh et al. (2017) captured the snap-off process during imbibition using fast synchrotron X-ray CT and stated the process was favorable for a large aspect ratio of local pores. Kuang et al. (2020) argued that wettability reversal after SI of the nanofluids was the main mechanism for the EOR based on the direct contact angle measurement in the pore space. The recent advent of in-situ CT has made it possible to directly image the rock and fluid distribution within the pores at the reservoir temperature and pressure (AlRatrout et al., 2018). However, such an experiment is expensive and the temporal resolution is insufficient to capture the detailed dynamic process during the SI.

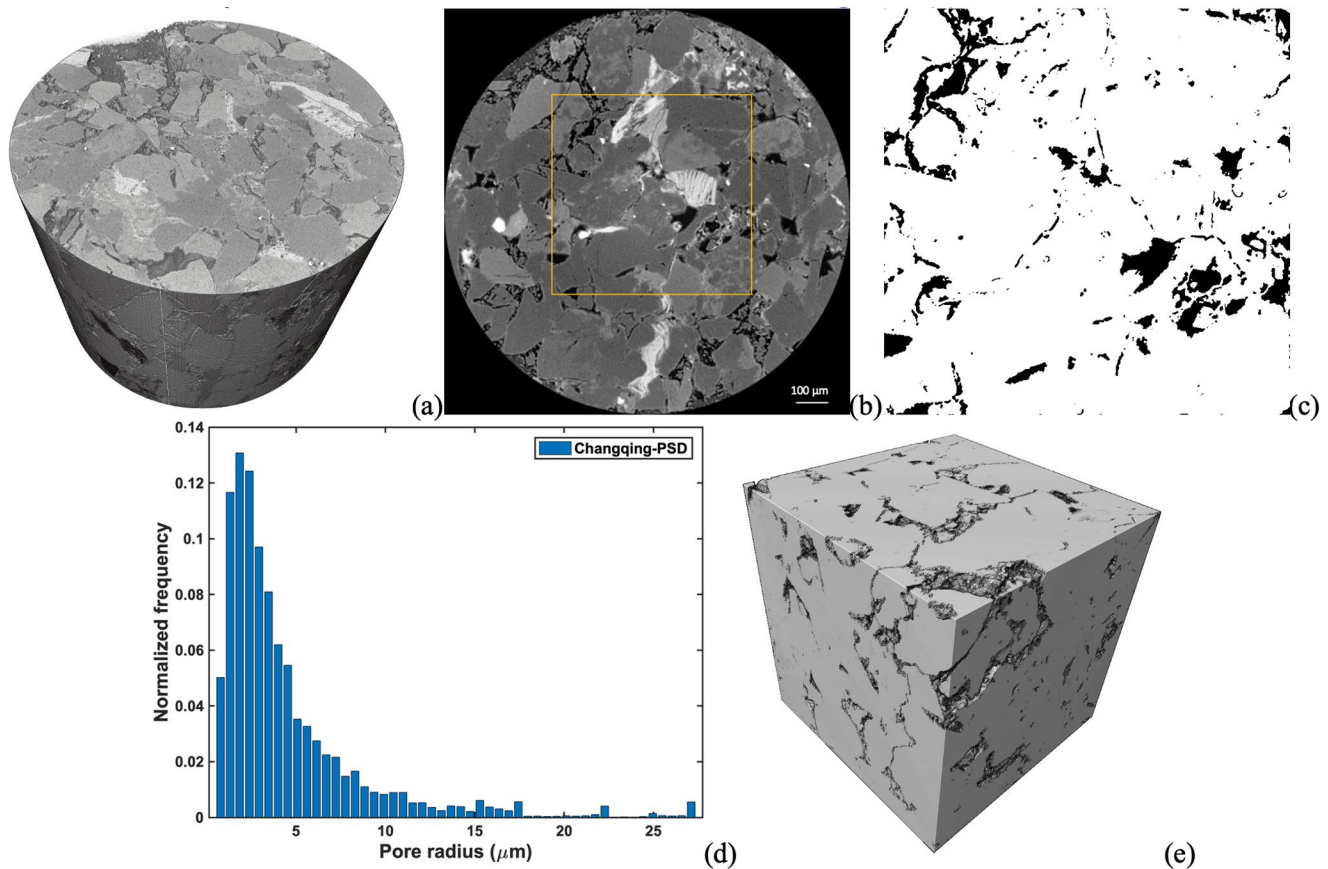
Several simulation methods can be used to investigate the multiphase flow dynamics in complex porous structures, such as computational fluid dynamics modeling (Jafari et al., 2017), pore network modeling (Qin et al., 2021; Valvatne & Blunt, 2004; Xie et al., 2017), and lattice Boltzmann method (LBM) (Ju et al., 2020; Li et al., 2016; H. Wang et al., 2019; Xie, Lei, & Wang, 2018; Xie, Lv, & Wang, 2018). Among them, the LBM, which is built upon the mesoscopic kinetic equations, stands out for modeling cases with complex boundary conditions and multiphase interfaces (S. Chen and Gary, 1998). There are four basic multiphase LBMs, which include the color gradient model, the pseudo-potential lattice Boltzmann method (SC-LBM), the free-energy model, and the phase-field model (Aidun & Clausen, 2010). Such direct pore-scale simulations of the SI in 3D porous structures enable one to capture the distinct influence of 3D pore morphology, pore surface wettability, and fluid properties on the imbibition dynamics. Liu et al. (2020) simulated the counter-current SI by the color gradient LBM and indicated the micro-fractures improved the oil recovery. Bakhshian et al. (2020) investigated the SI dynamics in fractured rock samples and argued the complex pore space geometries led to a non-uniform advancement of the interface. Zacharoudiou et al. (2017) simulated the two-phase flow dynamics in micro-fluidics by the free energy LBM and compared with the experimental results. Liu and Zhang (2017) simulated the dynamic behavior of a confined droplet by the phase-field LBM. Zheng et al. (Zheng, Chen, et al., 2018; Zheng, Ju, & Wang, 2018) employed the single component multi-phase SC-LBM to investigate the interface dynamics and the SI behavior in complex 3D porous structure. The above mentioned numerical methods provide suitable tools to quantitatively describe the SI behavior at the pore-scale. Among them, the SC-LBM is one of the most popular multiphase flow methods due to its easy implementation.

There is no doubt investigations of the flow dynamics at pore-scale and under reservoir condition are crucial to uncovering the controlling mechanisms of the complex SI behavior in unconventional rocks. During the shut-in operation, the SI of the fracturing fluid takes place in the complex pore space of the reservoir rock in high temperature and pressure conditions. In this work, we employed the multi-component SC-LBM to investigate detailed SI behavior in the porous structure of a tight sandstone sample. A high-resolution 3D image of the rock sample was obtained by micro-CT scanning. Then image processing techniques were employed to extract the 3D porous structure. An improved multi-component SC-LBM was developed to investigate the SI behaviors therein. The SI under ambient and reservoir conditions were calculated and compared. Moreover, the detailed snap-off of oil droplet during the SI was analyzed.

## 2. Materials and Methods

### 2.1. Porous Structure of a Tight Reservoir Sandstone

The rock sample was drilled from Changqing oilfield (China) and characterized as a low-permeability tight sandstone. The reservoir in Changqing oil field usually has high oil saturation and has an average temperature of 80°C and pressure of 15 MPa (S. Yang, 2017). The viscosity of the in-situ crude oil is around 1 mPa·s. In order to investigate the detailed pore-scale SI behavior, 3D porous structure of the sample was first imaged by a micro-CT scanner (Sanying, China). Totally, 1,258 slices with an image size of 1,630 × 1,630 pixels



**Figure 1.** The porous structure of a tight reservoir sandstone. (a) Reconstructed 3D grayscale image based on the computer tomography (CT) scanning data, (b) a 2D slice in the middle of the sample, (c) middle slice of the segmented binary model with black represents pores and white represents rock matrix, (d) pore size distribution of the sample, (e) compressed  $100^3$  voxels 3D model for the SI simulation.

were reconstructed from the scanning data. The reconstructed 3D grayscale image has a voxel resolution of  $0.772 \mu\text{m}$ , as shown in Figure 1a. A middle slice image of the sample is shown in Figure 1b. As observed, the sample is composed of granular particles and irregular high aspect ratio interparticle pores. The pore bodies mainly connect with each other through slit-shaped pore throats.

Several image processing procedures were applied to extract the porous structure from the reconstructed 3D image. First, a central  $800^3$  cubic region was selected for further processing, as illustrated in the middle slice by a yellow box in Figure 1b. This  $800^3$  cubic region with a voxel resolution of  $0.772 \mu\text{m}$  contained most pore morphology information of the CT scan data and considered as representative of the tight sandstone. Then the non-local mean denoising algorithm (Buades et al., 2005) was employed to decrease the image noise. Moreover, a Hessian-based filtering technique (Voorn et al., 2013), which providing an efficient way for better extracting the slit-shaped pores, was used in the image segmentation process. The middle slice of the segmented binary model is shown in Figure 1c with a comparison of the region in the yellow box in Figure 1b. The pore size distribution was calculated based on the binary model (Blunt et al., 2013; Dong & Blunt, 2009), which showed a quick decreasing trend as the pore size increase, as shown in Figure 1d. The calculated mean pore radius is  $4.462 \mu\text{m}$ . The model was further compressed to a  $100^3$  cubic for the SI simulation, as shown in Figure 1e. This is a compromise for the efficiency of the SC-LBM simulation. Several image processing techniques were applied in the model compression to ensure the connectivity of the porous model, which include the image erode algorithm to ensure the accurate characterization of the slit-shape pores and the image label algorithm for extracting the connected pores (T. Liu et al., 2018).

## 2.2. Multi-Component SC-LBM

An improved multi-component SC-LBM, with modification of the interaction force between fluids and solids, was employed to simulate the SI behavior in the porous model. The general framework of the model is similar to previous work (Coon et al., 2014; Porter et al., 2012), which used D3Q19 square lattice with explicit forcing terms and the multiple relaxation time (MRT) method. For completeness, we briefly introduce the main framework of the model with an emphasis on the improved fluids and solids interaction forces. In the mesoscopic SC-LBM,  $k$  particle distribution functions are introduced to represent the multi-component fluids. The distribution function is evolved using the discrete Boltzmann equation

$$f_{\alpha}^k(\mathbf{x} + \mathbf{e}_{\alpha}\Delta t, t + \Delta t) - f_{\alpha}^k(\mathbf{x}, t) = \Omega_{\text{coll}}^k + \Omega_{\text{forces}}^k \quad (1)$$

where  $f_{\alpha}^k(\mathbf{x}, t)$  is the density distribution function of the  $k$ th component in the  $\alpha$ th direction and it streams on the lattice sites  $\mathbf{x}$  at time  $t$  with fixed 19 directional velocities  $\mathbf{e}_{\alpha}$ . The left-hand side describes the streaming step of the distribution functions and the right-hand side describes the momentum change in the distribution functions due to collision ( $\Omega_{\text{coll}}^k$ ), and other forces ( $\Omega_{\text{forces}}^k$ ). The collision operator is defined as

$$\Omega_{\text{coll}}^k = M^{-1} \Lambda^k M \left[ f_{\alpha}^{\text{eq},k}(\mathbf{x}, t) - f_{\alpha}^k(\mathbf{x}, t) \right] \quad (2)$$

where  $M$  is the transformation matrix (d'Humières, 2002) that maps the distribution function to its moments,  $\Lambda^k$  is diagonal relaxation matrix and defined as  $\text{diag}[s_c^k, s_e^k, s_{\varepsilon}^k, s_c^k, s_q^k, s_c^k, s_q^k, s_c^k, s_q^k, s_{\nu}^k, s_{\pi}^k, s_{\nu}^k, s_{\nu}^k, s_{\nu}^k, s_{\nu}^k, s_m^k, s_m^k, s_m^k]$ . In this study, we chose  $s_c^k = 1, s_e^k = 0.1, s_{\varepsilon}^k = 0.1, s_q^k = 1.6, s_{\pi}^k = 1.4, s_m^k = 1.98$  with  $k = 1$  and 2 (Wu et al., 2018).  $s_{\nu}^k$  is related with the kinematic viscosity as  $\nu^k = \frac{c^2}{3} \Delta t \left( \frac{1}{s_{\nu}^k} - 0.5 \right)$ .  $f_{\alpha}^{\text{eq},k}$  is the equilibrium distribution function and expressed as follows:

$$f_{\alpha}^{\text{eq},k}(\mathbf{x}, t) = w_{\alpha} \rho^k(\mathbf{x}, t) \left[ 1 + 3 \frac{\mathbf{e}_{\alpha} \cdot \mathbf{u}^{\text{eq}}}{c^2} + 9 \frac{(\mathbf{e}_{\alpha} \cdot \mathbf{u}^{\text{eq}})^2}{2c^2} - 3 \frac{(\mathbf{u}^{\text{eq}})^2}{2c^2} \right] \quad (3)$$

where  $w_{\alpha} = 1/3$  ( $\alpha = 0$ ),  $w_{\alpha} = 1/18$  ( $\alpha = 1-6$ ), and  $w_{\alpha} = 1/36$  ( $\alpha = 7-18$ ).  $\rho^k$  is the macroscopic fluid density.  $c = \Delta x / \Delta t$  is the ratio of lattice spacing  $\Delta x$  and time step  $\Delta t$ .  $\mathbf{u}^{\text{eq}}$  is effective velocity, which has the same expression as total velocity  $\mathbf{u}$ , when  $s_c^k = 1$  with  $k = 1$  and 2.

The forcing operator on the right hand of Equation 1 is defined as

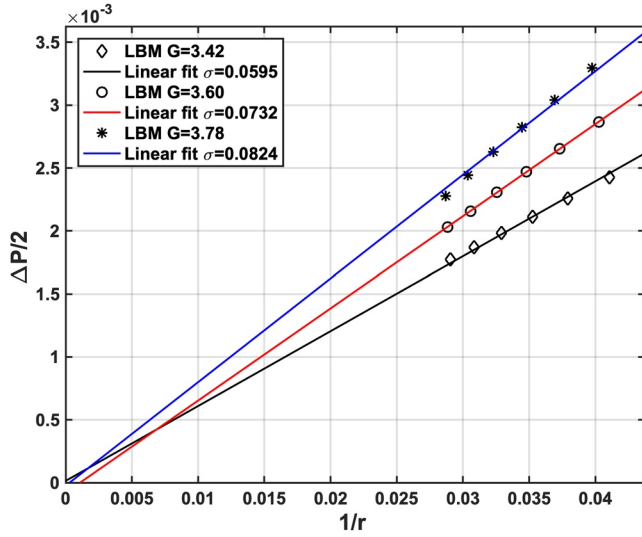
$$\Omega_{\text{forces}}^k = \frac{\Delta t}{2} \left[ f_{\alpha}^{F,k}(\mathbf{x} + \mathbf{e}_{\alpha}\Delta t, t + \Delta t) - f_{\alpha}^{F,k}(\mathbf{x}, t) \right] \quad (4)$$

where

$$f_{\alpha}^{F,k} = \frac{3\mathbf{F}^k \cdot (\mathbf{e}_{\alpha} - \mathbf{u}^{\text{eq}})}{\rho^k c^2} f_i^{\text{eq},k} \quad (5)$$

A transformation  $f_{\alpha}^{-k} = f_{\alpha}^k - \frac{\Delta t}{2} f_{\alpha}^{F,k}$  (Coon et al., 2014) is applied to Equation 1 to yield an explicit expression as

$$f_{\alpha}^{-k}(\mathbf{x} + \mathbf{e}_{\alpha}\Delta t, t + \Delta t) - f_{\alpha}^{-k}(\mathbf{x}, t) = M^{-1} \Lambda^k M \left[ f_{\alpha}^{\text{eq},k}(\mathbf{x}, t) - f_{\alpha}^{-k}(\mathbf{x}, t) - \frac{\Delta t}{2} f_{\alpha}^{F,k} \right] + \Delta t f_{\alpha}^{F,k} \quad (6)$$



**Figure 2.** Calculation of the surface tension with different values of  $G$ . The lines represent the least-square-fit of the simulation results.

The macroscopic properties for density, momentum, and total velocity are defined as follows

$$\rho^k = \sum_{\alpha} \bar{f}_{\alpha}^k \quad (7)$$

$$\rho^k \mathbf{u}^k = \sum_{\alpha} \bar{f}_{\alpha}^k \mathbf{e}_{\alpha} + \frac{\Delta t}{2} \mathbf{F}^k \quad (8)$$

$$\mathbf{u} = \frac{\sum_k \rho^k \mathbf{u}^k}{\sum_k \rho^k} \quad (9)$$

The forcing terms  $\mathbf{F}^k$  used in this work mainly include fluid-fluid inter-particle forces  $\mathbf{F}_{\text{int}}^k$  and the fluid-solid adhesive forces  $\mathbf{F}_{\text{ads}}^k$ , which control the surface tension and pore surface wettability in the simulation, respectively.

For the  $\mathbf{F}_{\text{int}}^k$  we only consider the interaction between different components which is defined as

$$\mathbf{F}_{\text{int}}^k(\mathbf{x}, t) = -G \rho^k(\mathbf{x}, t) \sum_{\alpha} w_{\alpha} \bar{\rho}^{\bar{k}}(\mathbf{x} + \mathbf{e}_{\alpha} \Delta t, t) \mathbf{e}_{\alpha} \quad (10)$$

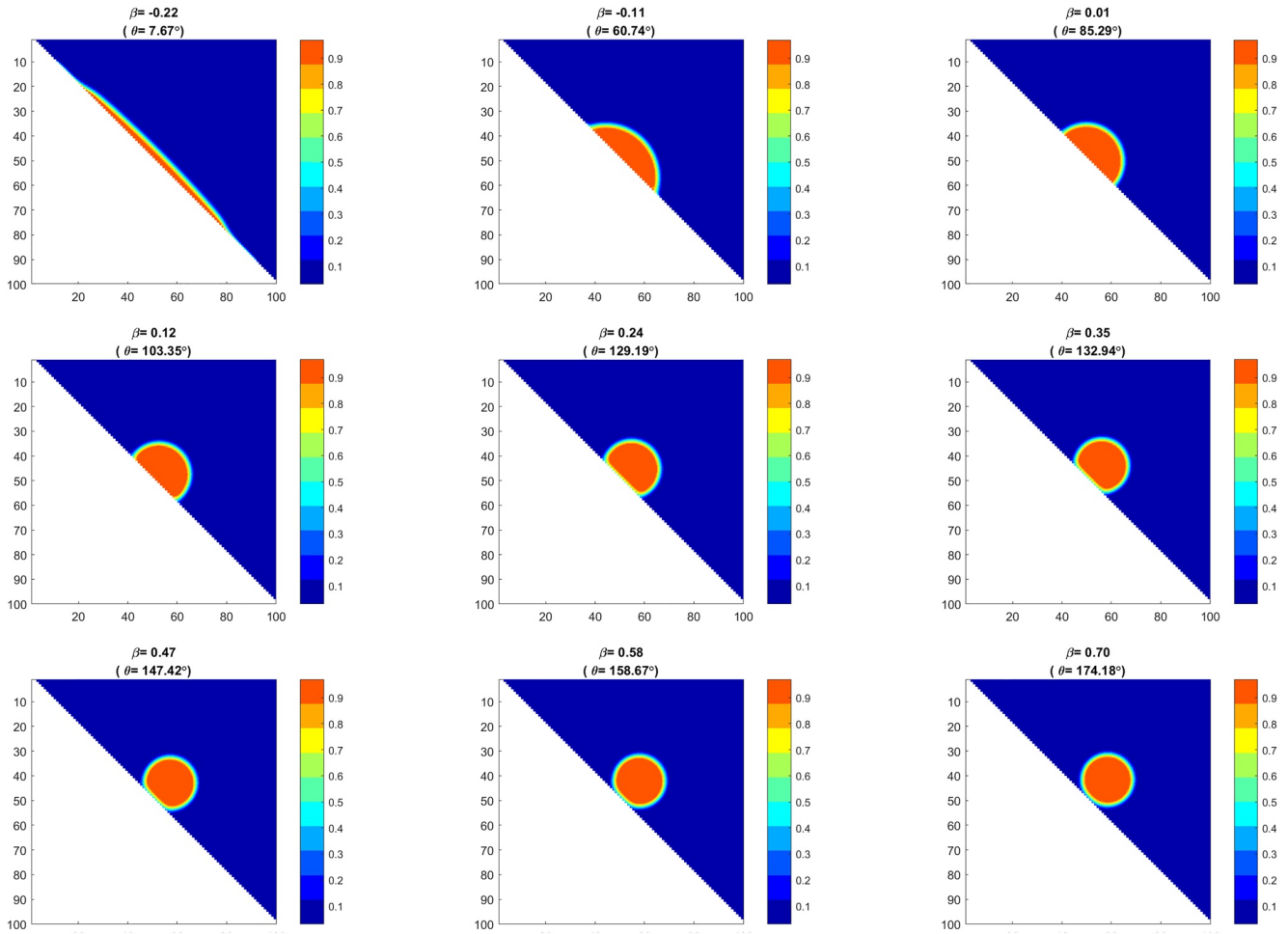
where  $G$  controls the strength of inter-particle forces and a large value of  $G$  permits the formation of an interface between different components. It should be noted, the multicomponent SC-LBM results in partially miscible fluids with each fluid node occupied by 1 and 2 components. For each fluid node away from the interface, one component is dominant ( $\rho_B$ ) and the other is considered as a dissolved minor component ( $\rho_S$ ). A larger value of  $G$  corresponds to larger surface tension and lower miscibility of the binary mixture. As presented by Porter et al. (2012), the surface tension obtained from 2D calculations with the explicit forcing term showed a quadratic relationship with the interaction strength. In addition, the obtained surface tension was independent of the viscosities of the components with the explicit forcing term. Here we calculated the surface tension with different values of  $G$  in a 3D domain. In each calculation, a spherical droplet of component 1 with initial radius  $R$  ranges from 25 to 35 lattices was placed in the center of a  $101 \times 101 \times 101$  lattice domain and the other nodes were occupied by the component 2. After reached equilibrium, the droplet radius  $r$  and the pressure difference  $\Delta P$  between the inside and outside the droplet were calculated. According to the Laplace law for a 3D case, i.e.,  $\Delta P / 2 = \sigma / r$ , the  $\Delta P / 2$  should have a linear relationship with  $1 / r$ . As shown in Figure 2, the calculated results agree well with the Laplace law.

Several studies have investigated the fluid-solid adhesive force  $\mathbf{F}_{\text{ads}}^k$  and its relation with the contact angle (L. Chen et al., 2014; Huang et al., 2007; Kamali et al., 2011; Porter et al., 2012). In general, the  $\mathbf{F}_{\text{ads}}^k$  was calculated through setting a constant fictitious wall density or a constant interaction strength between fluid and solid nodes. Such a procedure was easy to implement but would lead to unphysical density accumulation near the solid nodes, especially in a capillary force dominated SI simulation (De Maio et al., 2011).

In our previous studies (Zheng, Chen, et al., 2018; Zheng, Ju, & Wang, 2018), a modified fluid-solid interaction was presented for single component multiphase SC-LBM, which considered the fictitious wall density as a variable rather than a constant. Here, a modified  $\mathbf{F}_{\text{ads}}^k$  is employed to better control the fluid density accumulation for the multi-component SC-LBM. The basic idea is to eliminate the density accumulation near the solid nodes by controlling the interaction forces between the fluid and solid nodes. The  $\mathbf{F}_{\text{ads}}^k$  is defined as

$$\mathbf{F}_{\text{ads}}^k(\mathbf{x}, t) = -G \rho^k(\mathbf{x}, t) \sum_{\alpha} w_{\alpha} \bar{\rho}_w^{\bar{k}} S(\mathbf{x} + \mathbf{e}_{\alpha} \Delta t, t) \mathbf{e}_{\alpha} \quad (11)$$

where  $S(\mathbf{x} + \mathbf{e}_{\alpha} \Delta t, t)$  is an index function, which equals 1 for the solid nodes and 0 for the fluid nodes. The  $\bar{\rho}_w^{\bar{k}}$  is the fictitious wall density of the  $\bar{k}$ th component and its value is correlated with the average density of



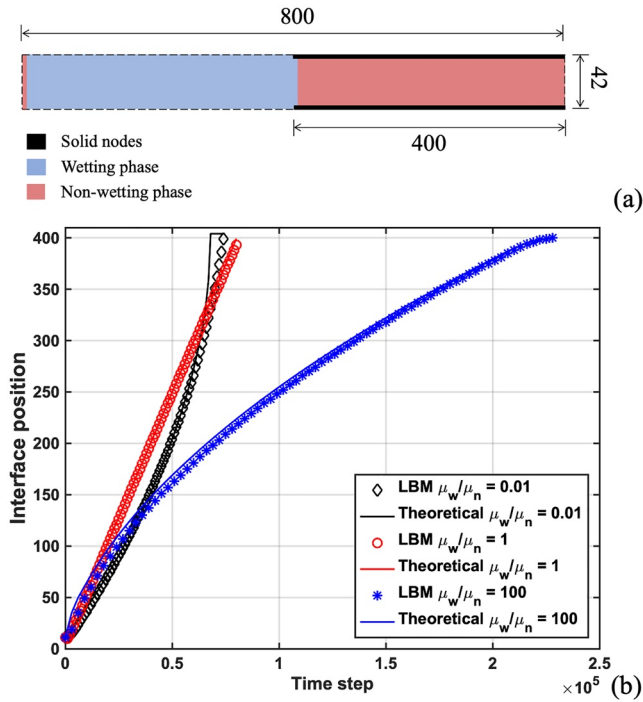
**Figure 3.** Calculation of the contact angle with different values of  $\beta$ .

its 26 neighboring  $k$ th fluid density ( $\overline{\rho^k}$ ). The value of  $\overline{\rho_w^k}$  is set as  $\rho_B$  when  $\overline{\rho^k} < 2\rho_S$ . Otherwise the value of  $\overline{\rho_w^k}$  is set as  $\beta$ . Similarly, the value of  $\rho_w^k$  is determined by the average density of its 26 neighboring  $\bar{k}$ th fluid density ( $\overline{\rho^{\bar{k}}}$ ). The value of  $\rho_w^k$  is set as  $\rho_B$  when  $\overline{\rho^{\bar{k}}} < 2\rho_S$ . Otherwise, the value of  $\rho_w^k$  is set as  $\rho_S$ . With different values of  $\beta$ , a contact angle ranges from nearly  $0^\circ$  to  $180^\circ$  can be accomplished.

Considering that the real pore surface is represented by zigzag lattice nodes, as shown in Figure 1e, we evaluated the contact angles with different values of  $\beta$  on a stair-shaped rough surface. The squared calculation domain has a size of  $N_x \times N_y \times N_z = 100 \times 1 \times 100$  lattices, which was divided by a diagonal line with the bottom-left region set as solid nodes and the up-right region set as fluids nodes. In each calculation, a semicircle droplet with a radius of 15 lattices of component 1 was initialized on the rough surface and the other fluid nodes initialized as component 2. The contact angle was calculated based on the interface profile after equilibrium. As shown in Figure 3, contact angles from  $7.67^\circ$  to  $174.18^\circ$  can be obtained with  $\beta$  values from  $-0.22$  to  $0.70$ .

### 2.3. Validation of SI in a Straight Channel

Considering a horizontal straight plate channel initially filled with the nonwetting phase, the SI naturally happened when exposing one end of the channel into the wetting phase. The process can be considered as quasi-static state with force balance between the capillary force and viscous force except for the very short early stage (Fries & Dreyer, 2009). Such a balance can be described by the following equation



**Figure 4.** Validation of three spontaneous imbibition (SI) cases in a straight channel. (a) Illustration of the SI simulation model, (b) comparison of the SI front propagation between the simulated and theoretical results.

$$\frac{2\sigma\cos\theta_a}{H} = \frac{12(dl/dt)}{H^2} [\mu_w l + \mu_n(L-l)] \quad (12)$$

where  $H$  and  $L$  are the width and length of the channel respectively,  $l$  is the advancing length of the wetting phase in the channel,  $\theta_a$  is the advancing contact angle,  $\mu_w$  and  $\mu_n$  are the viscosity of the wetting phase and the nonwetting phase, respectively. Let  $A = \mu_w - \mu_n$ ,  $B = \mu_n L$ ,  $C = H\sigma\cos\theta_a / 6$ , the Equation 12 simplified as:

$$\frac{dl}{dt} = \frac{C}{Al + B} \quad (13)$$

According to Equation 13, the velocity of the imbibition front ( $\frac{dl}{dt}$ ) would show a constant value when  $\mu_w = \mu_n$ , an increasing trend when  $\mu_w < \mu_n$ , and a decreasing trend when  $\mu_w > \mu_n$ . A general solution of Equation 13 is as follows by considering the initial length of the wetting phase is zero.

$$\frac{1}{2}Al^2 + Bl = Ct \quad (14)$$

Let us consider two specific conditions. First, when the viscosity of the nonwetting phase can be neglected compared to that of the wetting phase, that is,  $\mu_n = 0$  and  $B = 0$ , the Equation 14 simplified to the LW type equation as:

$$l = \sqrt{\frac{2C}{A}t} \quad (15)$$

which indicates that the interface front position is proportional with  $\sqrt{t}$ . It is easy to understand as that the driving capillary force keeps nearly constant but the resistant viscous force increases along the SI process.

Second, when  $\mu_w = \mu_n$  and  $A = 0$ , the Equation 14 simplified to

$$l = \frac{C}{B}t \quad (16)$$

which indicates that the wetting phase propagates linearly with  $t$ . The mechanism behind the linear behavior can be easily understood since the capillary driving force and total viscous resistance force remain a constant during the process.

The improved multi-component SC-LBM was used to simulate the SI in a straight channel. As illustrated in Figure 4a, the calculation domain has a size of  $N_x \times N_y \times N_z = 800 \times 1 \times 42$  lattices with the channel located on the right half of the model. The bounce-back boundary condition was implemented on the channel walls and the periodic boundary condition was implemented on the other boundaries. The wetting phase with a length of 400 lattices has 10 lattices overlap with the channel and the other fluid nodes were initialized with the nonwetting phase. The  $\beta$  value was set as  $-0.22$  to simulate a nearly completely wetting case. Three distinct SI cases were simulated with viscosity ratios  $\frac{\mu_w}{\mu_n} = 0.01, 1, \text{ and } 100$ . For the cases of  $\frac{\mu_w}{\mu_n} = 0.01$  and  $1$ , the  $s_v^2$  values were set equal to 1, and the  $s_v^1$  were set equal to 1.98 and 1, respectively. For the case of  $\frac{\mu_w}{\mu_n} = 100$ , the  $s_v^1$  was set equal to 1, and the  $s_v^2$  was set equal to 1.98. Comparison between the simulation and theoretical results are plotted in Figure 4b, which show good agreements. When  $\frac{\mu_w}{\mu_n} = 0.01$ , the imbibition showed an accelerating trend. When  $\frac{\mu_w}{\mu_n} = 1$ , a linear trend was observed as indicated by Equation 16. Moreover,

the imbibition trend was well described by the LW law, that is, Equation 15, when  $\frac{\mu_w}{\mu_n} = 100$ . These results validated the improved multicomponent SC-LBM in simulation the SI with different fluid properties.

### 3. Results

The co-current SI behaviors in an actual 3D pore space, as shown in Figure 1e, were simulated using the proposed multi-component SC-LBM under ambient and reservoir conditions, respectively. From here and after, the ambient condition represents that the temperature equals to 20°C and pressure equals to 0.1 MPa. The reservoir condition represents that the temperature equals to 80°C and pressure equals to 15 MPa. Comparisons of the SI behaviors under the two conditions were provided based on the simulation results.

#### 3.1. Model Setup

In this study, we chose pure water and n-decane as representative aqueous and oil phases, respectively. According to the data from the National Institute of Standard and Technology (<https://webbook.nist.gov/chemistry/>), the viscosity of water at the ambient condition and reservoir condition is 1.002 and 0.358 mPa s, respectively. The viscosity of n-decane at the ambient condition and reservoir condition is 0.913 and 0.523 mPa s, respectively. As a result, the viscosity ratio between the wetting phase and the non-wetting phase equals 1.097 at ambient condition and 0.685 at reservoir condition. The water and n-decane are far from their critical point at both conditions. At both conditions, the water and n-decane showed comparable viscosities.

Numerous works have been performed to measure the surface tension of water-hydrocarbon systems at different temperature and pressure conditions (Firoozabadi & Ramey, 1988; Georgiadis et al., 2011; McCaffery, 1972; S. Yang, 2017; Zeppieri et al., 2001). It was found that the surface tension between water and pure liquid hydrocarbon decreased with the temperature at a constant pressure. The change of surface tension with pressure could be neglected in most cases. The surface tension between water and n-decane is around 52 mN/m at ambient condition and 45 mN/m at reservoir condition according to the work of Georgiadis et al. (2011).

Moreover, it is found that temperature and pressure have little effect on the wettability of the oil-bearing reservoir rock (S. Yang, 2017). Here, we considered the pore surface is hydrophilic and the same wettability of the pore surface is set under the ambient and reservoir conditions. Compared with the ambient condition, the high temperature and pressure under the reservoir condition led to a smaller capillary force due to the slightly smaller surface tension as well as a smaller viscous force due to the smaller viscosity. In other words, the driven force and resistant force are both smaller under the reservoir condition than under the ambient condition. Thus, the SI behavior in these two conditions needs to be further investigated.

To quantify the SI behavior in the representative porous model under ambient and reservoir conditions, two parallel simulations were conducted with representative parameters for the two conditions. The calculation domain has a size of  $N_x \times N_y \times N_z = 150 \times 100 \times 100$  lattices, with the  $100^3$  size porous model located on the right of the domain. Initially, a 45 lattices long wetting phase plug was set in the left of the porous model with five lattices overlap. The bounce-back boundary condition was implemented on the solid nodes and the periodic boundary condition was implemented on the other boundaries.

In our previous simulation of liquid-gas SI case, where the gas viscosity can be safely ignored (Zheng, Ju, & Wang, 2018), we have shown an easy way to convert the lattice units into physical units by the Buckingham  $\pi$  theorem. Here liquid-liquid SI cases were calculated and one more  $\Pi$  term should be included, which can be chosen as the viscosity ratio between the wetting phase and the nonwetting phase. In the simulation, the viscosity ratios between the wetting phase and the nonwetting phase were set the same as that of the water and n-decane under the two conditions. In addition, the percentage of surface tension decrease from ambient condition to reservoir condition was also set according to that of the water and n-decane. Moreover, the pore surface was set as hydrophilic with a contact angle of 7.67° at both ambient and reservoir conditions. Parameters used in the two cases are list in Table 1.

**Table 1**  
Parameters Used in the SI Simulation Under Ambient and Reservoir Conditions

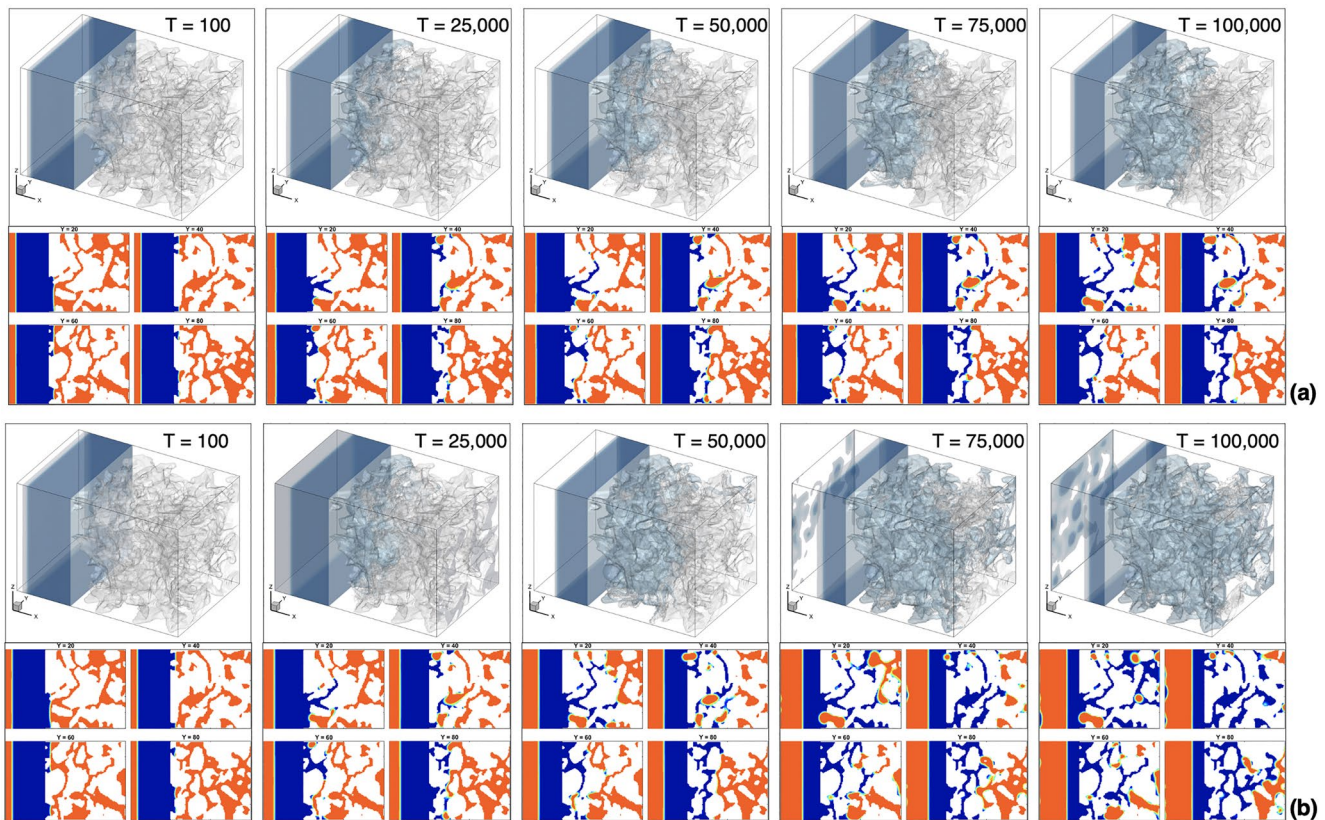
Cases	Ambient condition	Reservoir condition
$\rho_B$	0.97	
$\rho_S$	0.03	
$\beta$	-0.22	
$\theta$	7.67°	
$s_v^1$	0.67	1.16
$s_v^2$	0.71	0.98
$\mu_v^1$	0.33	0.12
$\mu_v^2$	0.30	0.17
$G$	3.60	3.47
$\sigma$	0.073	0.063

### 3.2. SI Behavior Under Ambient and Reservoir Conditions

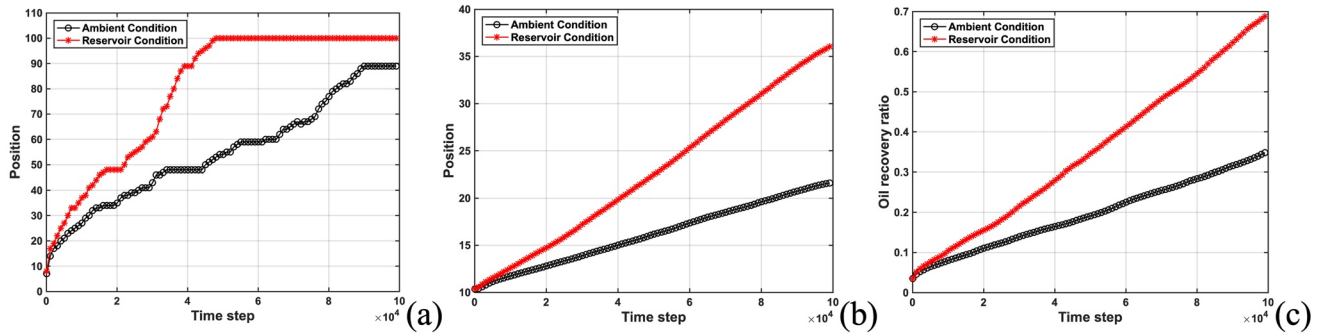
The SI behaviors in the representative porous structure of tight sandstone were simulated with the above settings for ambient and reservoir conditions. In both cases, the wetting phase imbibed into the model from the left surface and drove the nonwetting phase out the model from the right surface. A total time step of 100,000 was calculated for both conditions.

The SI pattern at different time steps under ambient and reservoir conditions are shown in Figures 5a and 5b, respectively. In general, the wetting phase imbibed quicker under the reservoir condition than the ambient condition. Part of the wetting phase front gradually reached to the right surface of the model under the reservoir condition, while none of the wetting phase fronts reached the right surface at the end of the 100,000 time step calculation. This was caused by the fact that the overall resistant viscous force under the reservoir condition was about half as much as that of the ambient condition, although the driving capillary force was slightly smaller.

The imbibition front of the wetting phase showed a complex morphology, which was usually named interface front roughening (Gruener et al., 2012; Qin et al., 2021). This phenomenon mainly occurred in high aspect ratio pores during the SI of fluids with comparable viscosity.



**Figure 5.** Visualization of the spontaneous imbibition (SI) behaviors in the representative porous model at the time steps of 100, 25,000, 50,000, 75,000, and 100,000. (a) Under the ambient condition, (b) under the reservoir condition. The upper row of each figure displays the 3D wetting phase morphology in the porous model, with the blue represents the water phase and the gray represents the pore space. The lower row displays the two phases distribution in the slices of  $Y = 20, 40, 60,$  and  $80$  of the porous model with the blue represent the water phase and the red represent the oil phase.



**Figure 6.** Calculation of the spontaneous imbibition (SI) pattern based on the simulation results. (a) Front tip position of the wetting phase, (b) tail interface position of the wetting phase plug, (c) oil recovery ratio in the porous model along with the SI.

Figure 6a shows the front tip positions of the wetting phase along with the SI for both conditions. In general, the front tip presented an irregular propagation pattern and the front tip propagated faster under the reservoir condition. Temporary stagnation of the front can be observed in both conditions, which occurred as the front meniscus propagating from the pore throat to the pore body.

Besides, we calculated the tail interface position of the wetting phase plug, as shown in Figure 6b. The tail interface propagation under the reservoir condition was about two times as fast as that under the ambient condition. For the reservoir condition, the tail interface position of the wetting phase plug showed a slightly accelerating trend along with the time step because a lower viscosity wetting phase was displacing a relatively larger viscosity nonwetting phase.

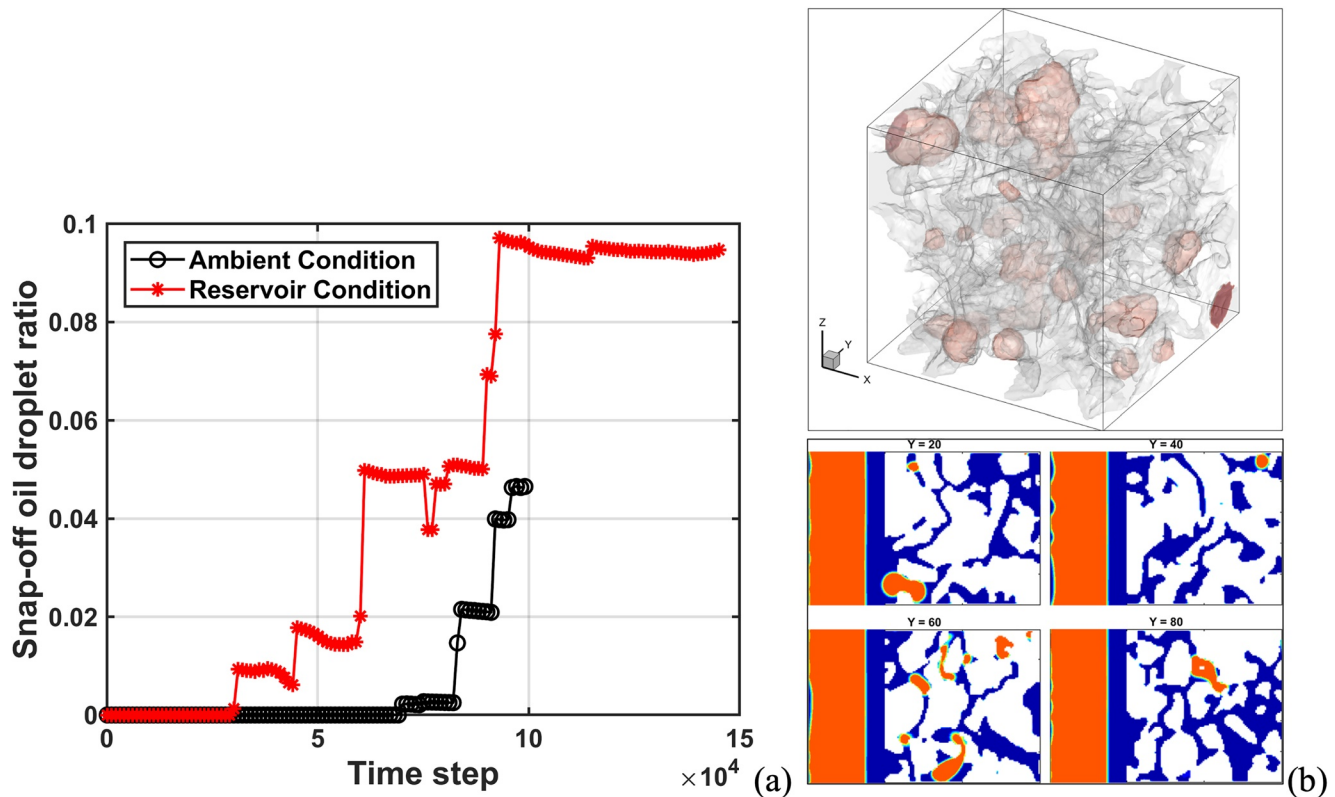
Moreover, we calculated the oil recovery ratio in the porous model, as shown in Figure 6c. As expected from the continuity hypothesis, the oil recovery ratio showed a very similar trend with the tail interface position. The oil recovery ratio reached 68.76% under the reservoir condition at the time step of  $T = 100,000$ .

### 3.3. Snap-Off of the Oil Droplets

Snap-off of the oil droplets can be observed under both conditions, as shown in Figures 5a and 5b, which usually happened in a pore body surrounded by slit-shaped pore throats. Such a snap-off behavior can hardly be captured by simplified models such as the capillary bundle model.

To quantify the snap-off behavior during the SI, we calculated the volume ratio of the snap-off oil droplets along with the simulations. As shown in Figure 7a, the occupied pore volume ratio of the snap-off oil droplets increased along with the SI. The curves showed jump increases when a new snap-off of the oil droplet occurred. A small decreasing trend of the curve can be observed after each jump. This was caused by the inherent compressibility effect of the SC-LBM. A rapid decrease of the snap-off oil droplet ratio occurred at  $T = 78,000$ . This was caused by the re-coalescence of the snap-off oil droplet with the front main oil plug. When a large portion of the water plug reached to the right surface of the model from  $T = 75,000$  to  $T = 100,000$  under the reservoir condition, several oil droplets were formed and stuck in the porous model, as shown in Figures 5b. This caused the rapid rise of the snap-off oil droplet ratio under the reservoir condition. Due to the slower SI under the ambient condition, the snap-off oil droplet ratio showed a slower increasing trend. The snap-off of the oil droplets accounted for parts of the trapped oil in an oil field. Moreover, these stuck oil droplets may block the transport channels and detrimental to further oil production.

In order to quantitatively characterize the snap-off of the oil droplet after the complete SI, we continuously calculated the SI in the porous structure under the reservoir condition. Further SI was observed in the model. At the time step around 146,000, the distribution of the water and oil phases reached an equilibrium and the SI nearly stopped in the porous model. As shown in Figure 7b, the oil droplets mainly stuck in the pore body. We labeled the oil droplets in the pore space by image processing methods. As calculated, 18 oil droplets, whose volume was larger than 100 voxels, dispersed and stuck in the pore body. These oil droplets occupied 9.47% of the pore space in the model and retarded the further flow therein due to the Jamin effect (Xu et al., 2019).



**Figure 7.** Snap-off of the oil droplets during the spontaneous imbibition (SI). (a) Occupied pore volume ratio of the snap-off oil droplet along with the SI. (b) Final distribution of the water and oil phases in the porous model under the reservoir condition. The upper row displays the oil droplet in the porous model, with the red represent the oil droplet and the gray represent the pore space. The lower row displays the two-phase distribution in the slices of  $Y = 20, 40, 60,$  and  $80$  of the porous model with the blue represents the water phase and the red represents the oil phase.

#### 4. Conclusions

An improved multi-component SC-LBM was developed with the modification of interaction force between the fluid and solid nodes. Good agreement between the simulation and theoretical results of different viscosity ratio SI cases in a straight channel demonstrated the ability of the proposed method in simulating the SI under various conditions. We employed the proposed method to simulate the SI behavior in a representative porous model under the ambient and reservoir conditions. Comparison of the SI in the same porous model showed that the imbibition of the wetting phase under the reservoir condition is about two times as fast as that under the ambient condition. The main reason caused the faster SI under the reservoir condition is the lower overall resistant viscous force. Besides, the snap-off of the oil droplets during the SI mainly occurred in a pore body surrounded by slit-shaped pore throats. The snap-off caused the oil droplets stuck in the pore space. At the end of the SI process under the reservoir condition, dispersed and stuck oil droplets accounted for 9.47% of the pore volume. The distinct SI behavior under the ambient and reservoir condition suggested that the laboratory SI data, which usually obtained under ambient condition should be carefully applied in interpreting the underground cases. Moreover, the snap-off of the oil droplet in the complex pore space indicated that the simplification of the pore space into simple geometries would cause a discrepancy in predicting the water SI into the oil filled complex pore structures, especially those with high aspect ratio.

#### Data Availability Statement

The data for this study can be obtained from <https://figshare.com/s/0c9e5de9f7484728c75c>.

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