

## Invited review

## Spontaneous imbibition in shale: A review of recent advances

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shale-liquid interaction**Cited as:**Li, C., Singh, H., Cai, J. Spontaneous imbibition in shale: A review of recent advances. *Capillarity*, 2019, 2(2): 17-32, doi: 10.26804/capi.2019.02.01.**Abstract:**

Spontaneous imbibition in shale is commonly observed during and after hydraulic fracturing. This mechanism greatly influences hydrocarbon recovery in shale plays, therefore attracting increasing attention from researchers. The number of related publications is increasing. In this review, we summarize the recent advances in shale spontaneous imbibition from three aspects, namely, conventional spontaneous imbibition models, common experimental methods, and key mechanisms that need to be focused on. The major influencing factors on the shale imbibition process are discussed, and promising future works are presented on the basis of the merits and demerits of the recent studies. This study discusses the mechanisms of shale-liquid interaction, the complexity of multiphase flow in shale plays, and highlights the achievements and challenges in shale imbibition research.

**1. Introduction**

Shale reservoir rocks have relatively low permeability and connectivity in comparison to conventional reservoir rocks like sandstone and carbonate, therefore, to achieve economical production hydraulic fractures are created to increase matrix connectivity. To achieve this, several million gallons of fracturing liquid is injected at high pressure into shale plays (Palisch et al., 2010; Soliman et al., 2012) to create hydraulic fractures and proppants are mixed with the liquid to keep the fractures from closing. The injected fracturing liquid triggers a series of liquid-shale interaction processes that are affected by the operational conditions such as injection, flowback, shut-in, and production; spontaneous imbibition is one of such key liquid-shale interactions. Although spontaneous imbibition, in other words liquid uptake by the porous medium without an enforced pressure, is a universal phenomenon in porous media and its diverse applications (Singh and Myong, 2018), its occurrence in shale rocks is understood to a relatively lesser extent because of complex pore structure and heterogeneous properties. Production data shows that, generally for high production wells only 6%-10% of injected water can be recovered during the fracturing process (Vandecasteele et al., 2015) and up to 48% in the entire production history (Nicot et al., 2012; Roychaudhuri et al., 2013). A large amount of

fracturing liquid that is not recovered back is assumed to be imbibed by shale. The mechanisms of spontaneous imbibition in shale are complicated (Singh, 2016) and have attracted increasing attention in recent years.

The influence of spontaneous imbibition on shale production is another area about which not much is known. The liquid retained in shale formation causes formation damage under several circumstances (Bennion et al., 2005). A strong imbibition may absorb liquid retained in the main flow channel and unblock the channels during the shut-in period, recover permeability of blocked channels, increase reservoir pressure, and enhance oil/gas production under other circumstances (Wang et al., 2015; Shen et al., 2017). A series of laboratory observations (high-precision balances, nuclear magnetic resonance/NMR, X-ray micro/nanocomputed tomography and iron tracer discussed in section 2) and mathematical models (Handy's, capillary bundle, fractal, theoretical, and other related models discussed in section 3) have been reported in the past few years.

However, there is still a large gap in terms of verifying the laboratory-based observations at field-scale as well as their simulation by mathematical modeling. Therefore, reviewing the current state of research for spontaneous imbibition in shale and understanding its implication on production of fluids



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at field-scale is an important topic for research.

Some studies have previously reviewed imbibition (Cai and Yu, 2012; Mason and Morrow, 2013; Singh, 2016) in greater detail for both convention and unconventional reservoirs. These previous studies have mostly focused on providing a detailed overview of the mechanisms involved in driving the imbibition, whereas the focus of this study is to review the advancements in experimental and mathematical techniques in regards to the investigation of imbibition.

In this study, we review recent advances in spontaneous imbibition research for shale and summarize them into three parts. The first part discusses experimental advances, including recent methods, instruments, and observations. The second part discusses advances in mathematical modeling of imbibition, including the development of analytical, semi-analytical, and numerical simulations. The third part provides several factors influencing spontaneous imbibition in shale, and potential ideas for future research.

## 2. Experimental progress in laboratory methods for observation of spontaneous imbibition

Many studies have focused on the factors influencing spontaneous imbibition in shale (Nelson, 2009; Dehghanpour et al., 2012; Xu and Dehghanpour, 2014; Singh, 2016; Zhou et al., 2016; Javaheri et al., 2017; Siddiqui et al., 2018), including capillarity in organic and inorganic pore network systems, initial and induced fractures caused by liquid-clay mineral interactions, pore structure (pore size distribution) and microstructure (roughness, tortuosity and connectivity), wettability, osmotic forces, mineral contents, liquid-flowing behavior, and physical and electrochemical interactions between liquid and shale. Instead of reviewing studies on the basis of factors influencing liquid uptake by shale, the review in this section focuses on progress in laboratory methods and techniques to observe spontaneous imbibition. Experimental techniques in observing spontaneous imbibition have been reported by the use of nuclear magnetic resonance (NMR), micro and nano-computed tomography (CT), and balance-weights, which are elaborated in the following three subsections, respectively.

### 2.1 Nuclear magnetic resonance (NMR)

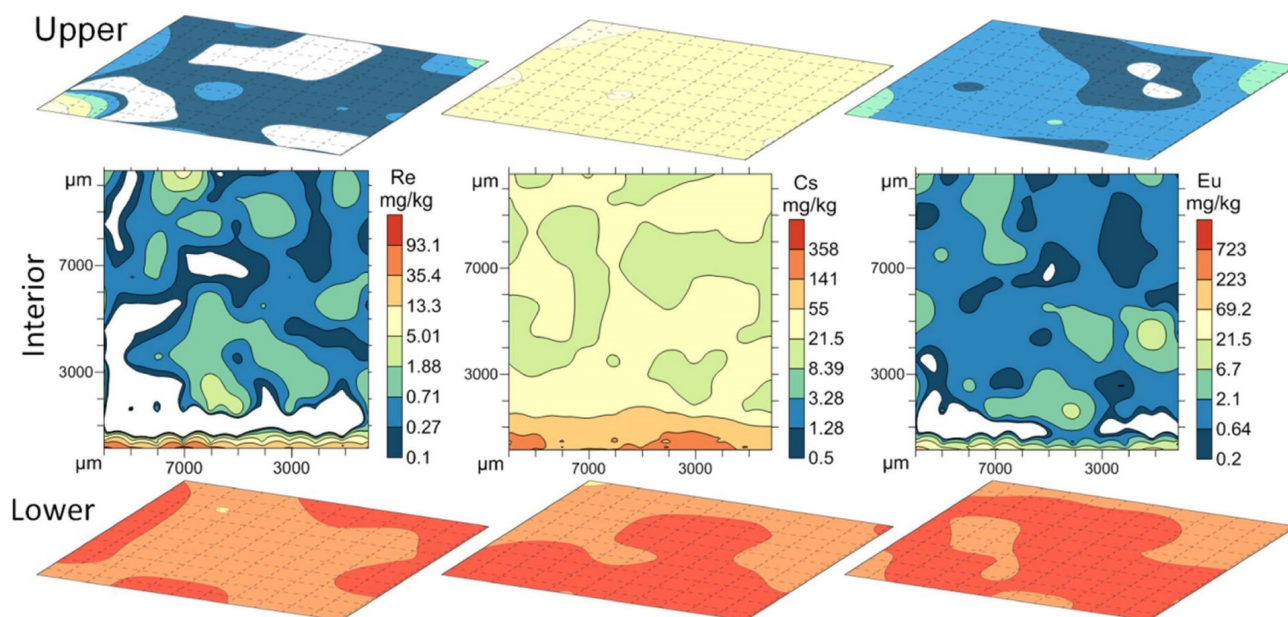
Nuclear magnetic resonance (NMR) can reflect the liquid distribution among different pores in a rock, which is a direct way to monitor the liquid dynamics as it imbibes in shale. It can monitor liquid-displaced gas and oil directly by imaging the liquid distribution in rocks. Meng et al. (2015) used NMR to monitor the Longmaxi Formation shale imbibition in co-current and counter-current imbibition. Results showed that the liquid first filled the small pores and then the large pores in both conditions. Small pores had strong imbibition force (Meng et al., 2015). Dong et al. (2015) used NMR to reflect liquid saturation established by imbibition. The NMR was used to monitor the sufficient and insufficient liquid imbibition to study the imbibition characteristics (Meng et al., 2016). Results showed that the liquid filled small pores preferentially during sufficient liquid imbibition, whereas it moved from

large pores to small pores during insufficient liquid imbibition. Liu et al. (2016a) used NMR to monitor insufficient imbibition and used a preservative film to wrap the imbibed sample. Liquid similarly moved from large to small pores. Lin et al. (2016) used NMR to investigate the imbibition-front progression of shale and found that liquid saturation gradually increased in samples. The water saturation was almost 100% near the imbibition face but low far away. Yang et al. (2018a) investigated sufficient liquid imbibition in shale. Liquid filled the small pores preferentially in marine shale and the large pores preferentially in continental shale. Liu and Sheng (2019) used NMR to study surfactant imbibition in shale and found surfactant-enhanced imbibition. Li et al. (2019) used NMR to study aqueous migration during well shut-in period and found that liquid migrated from large pore spaces to small ones. This phenomenon benefitted permeability recovery. Dai et al. (2019) used NMR to investigate surfactant imbibition to displace oil from shale. Surfactant displaced the oil in small pores preferentially. Zhou et al. (2019) used NMR to study the oil displacement process by nanofluid imbibition and found that nanofluids can enhance oil recovery. The above observations reported with the help of NMR show its effectiveness as a novel and robust method in studying spontaneous imbibition in shale.

### 2.2 Micro/nano-CT

With the development of imaging instruments and analyzing methods, microstructures in shale can be observed directly. A series of imaging methods is available, and each method has its special benefit and limitation. These methods include HIM (2D, common resolution 1 nm, only for organics), FIB (3D, resolution up to 5 nm), SEM (2D, resolution up to 4 nm), nano-CT (3D, resolution up to 65 nm), and micro-CT (3D, resolution up to 1  $\mu\text{m}$ , common for cracks) (Zhou et al., 2018). Zheng et al. (2018) briefly summarized the recent advances in these methods for their application in observing shale fluid-rock interaction. Several studies have also provided modeling methods that are based on the use of two or more of these experimental techniques, utilizing each method's superiority, for example using SEM to develop a 3D intermingled-fractal model (Li et al., 2017, 2018a). Chakraborty et al. (2017) used micro-CT imaging to observe shale microcracks introduced by liquid imbibition. Siddiqui et al. (2019) used micro-CT imaging to scan the internal structure of samples before and after spontaneous imbibition at unconfined and confined stress states. There have been other similar studies published in recent years, but almost no experimental technique can directly observe the spontaneous imbibition process in shale nanopores. Our research group have tried to use micro-CT to observe imbibition in shale directly with KI (potassium iodide) as a tracer, but the result is really bad. The resolution is insufficient for reconstruction and additional analysis.

Hu et al. (2012) provided an "indirect" method to observe forced-imbibition process and analyze the connectivity of pore space, especially in shale, and developed this method in recent years (2015a, 2015b). During the test, the shale sample was forcibly injected with a molten wood's metal alloy, which is



**Fig. 1.** Profiles of nonsorbing ( $\text{ReO}_4^-$ ) and sorbing tracers ( $\text{Cs}^+$  and  $\text{Eu}_3^+$ ) for API brine fluid imbibition into the shale sample after 12 hr (Yang et al., 2018b).

combined by 50% Bi, 26.7% Pb, 13.3% Sn, and 10% Cd, with melting point around  $78^\circ\text{C}$ . The sample was then cooled down to room temperature. The metal alloy solidified and occupied small cracks and matrix pores connected to the sample surface. Based on calculation, when injection pressure reaches 154 MPa (1542 bars), the pore-throats of diameter greater than 9.2 nm can be invaded (Hu et al., 2015b). With the use of SEM method, the connecting structures could be mapped by measuring the concentration of each component. Although the method originated from Swanson (1979) and Dullien (1981), the successful application on shale makes it an effective tool to analyze connectivity. Substantial progress has occurred on the basis of this method. Fig. 1 shows that the connectivity of pore space in shale can be directly observed by using the metal alloy imbibition method (Yang et al., 2018b; Yang et al., 2019). As illustrated in Fig. 1, anionic tracer  $\text{ReO}_4^-$  is nonsorbing, the profile may show distribution of small connecting cracks, and  $\text{Cs}^+$  is sorbing. The diffusion effect can be observed by comparing the difference in concentration profiles between sorbing and nonsorbing tracers. The ion tracing method has revealed several mechanisms facilitating spontaneous imbibition in shale to a certain extent.

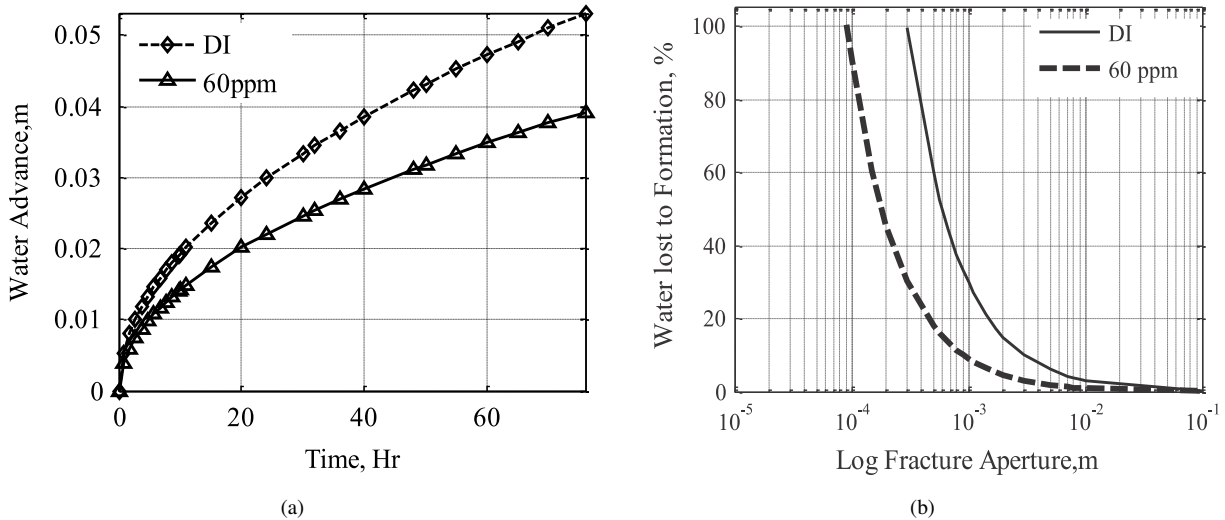
### 2.3 Balance-weighting methods

Balance-weighting technique is a traditional method to test a sample's imbibition behavior. It generally comprises of two methods, namely, all-submerged and surface-submerged methods. In the first method, the sample is submerged in liquid; as the sample imbibe liquid, the sample's weight increases. This method is commonly used to test counter-current imbibition. In the latter method, only the bottom face contacts liquid. As imbibition proceeds, the sample's weight

increases. Evaporation leads the height of the liquid surface and the submerged volume of sample to decrease; thus, the balance weight increases. For the surface-submerged method, an impermeable correction sample is needed to diminish the influence of evaporation. The surface-submerged method is commonly used for co-current imbibition. Detailed information can be found in many studies, like Shen et al. (2016, 2017).

Roychaudhuri et al. (2013, 2014) used a balance-based imbibition instrument to explore the role of capillarity in the fluid loss mechanism of shale plays. They used Marcellus shale cube with an edge length of 1 cm; only one face of the cube was exposed to water, and other five faces were covered with impermeable epoxy. The test simulated the counter-current imbibition process. They observed a distinct transition from initial imbibition rate, which depended linearly on the square root of time to a lower rate at later times. They used simple slab geometry to estimate water loss into the formation. They calculated that the front penetration distances between the cases with (60 ppm surfactant) and without surfactants (DI, deionized water) differed significantly, as shown in Fig. 2(a). The fractional fluid loss to the formation due to imbibition as a function of fracture aperture is discussed, as shown in Fig. 2(b).

Gao and Hu (2016) tested samples on balance in directions parallel and transverse to the lamination and in imbibing fluids of deionized water and n-decane to investigate the influence of shale layers. Mehana et al. (2018) investigated the influence of salinity and mineralogy on imbibition kinetics by using an all-face-open shale sample on a balance-based imbibition instrument. Recently, Roychaudhuri et al. (2019) performed forced imbibition experiments with shale samples from various depths in the Marcellus formation to investigate the influence



**Fig. 2.** The movement of imbibition front and the relationship between water loss and fracture aperture (Roychaudhuri et al., 2013).

of salinity on the petrophysical properties of shale during the flowback water reinjection process. They found that repeated forced imbibition and flow-back process can increase porosity, which is likely due to the creation of macrofractures and the mineral dissolution. However, the impact of permeability is more nuanced, which is controlled by the combined effect of additional porosity being created and the presence of residual water saturation/water blockage.

### 3. Modeling techniques for spontaneous imbibition

Although Singh (2016) summarized imbibition in shale a few years ago, new modeling studies have been published in recent years, taking an important step toward understanding the mechanism of multiphase displacement and spontaneous imbibition in shale. Below, we first briefly review the progress in classical modeling of spontaneous imbibition based on Lucas (1918) and Washburn (1921), followed by recent advances in modeling for imbibition in shale since the review by Singh (2016).

#### 3.1 Advances in Lucas and Washburn Equation

Since Lucas (1918) and Washburn (1921) derived the basic law of spontaneous imbibition in vertical capillaries and developed the Lucas-Washburn model (LW model), numerous experimental and theoretical studies have been developed based on this theory. Handy (1960) developed imbibition theory for homogeneous porous media and established classic Handy's model. In Handy's original model, wetting liquid is imbibed into porous media by capillary force and the imbibition process can be simplified as piston-like displacement. Thus, cumulative mass imbibed into the porous media varies linearly with the square root of imbibition time. For conventional sandstone, experiments have shown that imbibition behavior in sandstone can be described by Handy's model. Capillary force is the dominant driving force for spontaneous imbibition

in sandstone rocks, which have large and fairly homogeneous pores that can be simplified as bundles of capillary tubes (Rao et al., 1976; Nutt, 1982).

The LW equation and Handy's model have been further developed in the past decades. Martic et al. (2002) showed that the contact angle in the LW equation should be a dynamic angle instead of a constant variable. Fries and Dreyer (2008) introduced an inclined tube in place of the vertical tube in the original LW equation. Kim and Whitesides (1997) developed an imbibition equation for noncircular tube. Yu et al. (2002, 2009) included fractal dimensions in the original LW equation to account for tortuosity in capillary rise. Zhang et al. (1996) defined a characteristic imbibition length to correlate imbibition results for systems with different shapes and boundary conditions. Li and Horne (2000) provided an implicit expression for imbibition that include the effect of gravity. Other imbibition models have been summarized by Manson and Morrow (2013).

Besides the capillarity that drives spontaneous imbibition and mathematically represented by the LW equation, representing other mechanisms driving spontaneous imbibition in shale is not possible through LW equation. Here we review recent developments in modeling spontaneous imbibition using various methods, such as molecular dynamics simulation, lattice Boltzmann simulation, pore network modeling, and continuum-scale physics.

#### 3.2 Molecular dynamics (MD) simulation method

##### 3.2.1 Theory

MD is a simulation technique to understand the fluid flow and its interaction with the flowing medium at the molecular (and sub-molecular) level. MD simulation often serves as a complement to the conventional experiments as well as it can be used to realistically mimic a phenomenon that is sometimes difficult to conduct experimentally for different reasons. The reason why MD simulation is able to mimic an experiment is because no assumptions are made about

the process/mechanism that needs to be investigated, except the fact that the total energy (potential + kinetic) should be conserved.

MD simulation involves numerically solving the Newton's equations of motion to determine the trajectories of interacting particles and the force acting on a fluid particle is typically the sum of three components, which are i) the force due to interaction between the particle with other fluid particles, ii) force between the particle and the particles composing the solid wall, and iii) external force such as gravity, pressure gradient etc. Therefore, we can write the total force  $F(\vec{r}_i)$  on a fluid particle  $i$  as follow:

$$F(\vec{r}_i) = m_i a_i(t) = m_i \frac{d\vec{r}_i^2}{dt^2} \quad (1)$$

$$m_i \frac{d\vec{r}_i^2}{dt^2} = \underbrace{\sum_{j \neq i, j=1}^N F_1(\vec{r}_i, \vec{r}_j)}_I + \underbrace{\sum_{j_w \neq i, j_w=1}^{N_w} F_2(\vec{r}_i, \vec{r}_{j_w})}_{II} + \underbrace{F_3}_{III} \quad (2)$$

where, term *I* represents force due to interaction between fluid particle  $i$  and other fluid particles in the medium, term *II* represents the force between fluid particle  $i$  and all the particles of the solid wall, term *III* represents the external force such as gravity, pressure gradient etc. Here,  $m_i$ ,  $a_i(t)$ ,  $\vec{r}_i$  are the mass, acceleration and position coordinate of the particle  $i$  at time  $t$ . Force on the particle can be written as a gradient of the potential energy as shown:

$$F(\vec{r}_i) = -\frac{dE(\vec{r}_i)}{d\vec{r}_i} \quad (3)$$

where, the total potential energy  $E(\vec{r}_i)$  on particle  $i$  is given as following:

$$E(\vec{r}_i) = \underbrace{\sum_{j \neq i, j=1}^N \phi_1(\vec{r}_i, \vec{r}_j)}_I + \underbrace{\sum_{j_w \neq i, j_w=1}^{N_w} \phi_2(\vec{r}_i, \vec{r}_{j_w})}_{II} + \underbrace{\phi_3(\vec{r}_i)}_{III} \quad (4)$$

where, term *I* represents energy due to interaction between fluid particle  $i$  and other fluid particles in the medium, term *II* represents the energy between fluid particle  $i$  and all the particles of the solid wall, term *III* represents the energy due to external field such as gravity, pressure gradient etc. Potential energy between the fluid/fluid ( $\phi_1$ ) and fluid/wall ( $\phi_2$ ) particles can take different forms depending on whether the interactions between the particles are non-bonded or bonded (attractive or repulsive), and typically the potential energy for the former is calculated by the Lennard-Jones (LJ) potential (also known as the potential or the van der Waals potential) that is the sum of energy contributions between pairs of atoms. Whereas, in the case of bonded interactions between the particles such as in the presence of electrostatic charges, an additional term of Coulomb potential must be added to the LJ potential for the pair of charged particles.

$$\phi_1 = \phi_2 = \begin{cases} \phi_{LJ}, & \text{non-bonded interaction} \\ \phi_{LJ} + \frac{z_i z_j}{4\pi\epsilon_0 r_{ij}^2}, & \text{bonded interaction} \end{cases} \quad (5)$$

where,  $\phi_{LJ} = 4\epsilon \left[ \left( \frac{\sigma}{r_{ij}} \right)^{12} - \left( \frac{\sigma}{r_{ij}} \right)^6 \right]$ . Here,  $\phi_{LJ}$  is the energy due to LJ potential,  $\vec{r}_{ij} (= \vec{r}_i - \vec{r}_j)$  is the distance between two particles  $i$  and  $j$ ,  $\sigma$  is the particle length (or diameter for spherical particles),  $\epsilon$  is an interaction strength parameter,  $\epsilon_0$  is the permittivity of free space,  $z_i, z_j$  are the charges on the pair of particles  $i, j$ .

Other than the external forces on liquid particles, the particles within the solid walls of micro- and nano-channels also experience an external force in form of spring force that is typically employed to keep the particles of the solid wall oscillating about its lattice site (Fan et al., 2002) using a Hookean spring as follow:

$$\phi_{3,w} = \frac{H_1}{2} R^2 + \frac{H_2}{4d_{cr}^2} R^4 + \frac{H_3}{6d_{cr}^4} R^6 \quad (6)$$

where,  $H_1, H_2, H_3$  are the spring constants,  $R$  is the displacement of the wall particle from its lattice site, and  $d_{cr}^2$  is the critical mean-square displacement of wall particles that is determined as per the following Lindermann criterion (Hansen and McDonald, 2006)  $\frac{\langle R^2 \rangle}{d_{min}^2} < 0.023$ , where  $d_{min}$  is the distance to the nearest neighbor. This criterion requires  $d_{cr}^2 < 0.023 d_{min}^2$ , and therefore, typically it is assumed  $d_{cr}^2 = 0.01 d_{min}^2$ .

Because a set-up of MD simulation is complicated process, its applications are mostly limited to the fundamental study of simple geometry like plane wall, channel and cylinder, and it is commonly used to simulate flow at the micro/nanoscale.

### 3.2.2 Application towards imbibition in shale

Originally, on flowing behavior analysis, MD method is often used to analysis dynamics of water confined between silica surfaces and pores (Gallo et al., 2000, 2001; Bonnaud et al., 2010) and to analysis adsorption of water molecules in slit pores (Spohr et al., 1998). With the development of MD method and computational ability, liquid flow in nano tubes can be simulated. Supple and Quirke (2004, 2005) built a carbon nanotube and simulated the oil imbibition behavior by MD method. They found that a single wall carbon nanotube filled with a linear time dependence at short time, possibly caused by low friction of graphitic surface and the short time scale. For rougher surfaces, imbibition process followed the Washburn equation. Wang et al. (2016) also simulated oil transport through inorganic nanopores in shale by MD method. In their model, liquid hydrocarbon (octane) was driven through inorganic (quartz) nanopores ranging in size from 1.7 to 11.2 nm. Through MD simulation, they provided both slip length and apparent viscosity methods to correct liquid flow, and discussed influencing factors on slip length. Yang et al. (2017) simulated the dynamics of octane and water molecule imbibition into graphite and quartz slits with a width of 1-2 nm. Results showed that oil can imbibe into the organic nanopores of tight rocks faster than what classic imbibition models predict. Fast imbibition of octane may be caused by the higher relative density of octane compared with that of water in graphite slit. In summary, limited by basic calculation theory, the advantage of MD is the ability to investigate flowing behavior at the nanoscale. Upscaling the space, increasing the

molecule number, or extending the simulation time lead to enormous computational complexity.

### 3.3 Lattice Boltzmann (LBM) method

#### 3.3.1 Theory

In Lattice Boltzmann methods (LBM), the discrete version of kinetic Boltzmann (or Boltzmann with simplified collision models) equation defined on the finite lattice is solved to simulate fluid flow. The LBM models the evolution of a density distribution (together with internal energy density distribution in case of thermal flow) associated with fictive particles that perform consecutive movement and collision processes over a discrete lattice mesh. In LBM,

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f = \frac{f^{(0)} - f}{\tau} \quad (7)$$

is reduced to

$$f_i(t + \Delta t, r + v_i \Delta t) - f_i(t, r) = \frac{\Delta t}{\tau} [f_i^{(0)}(t, r) - f_i(t, r)] \quad (8)$$

By introducing a finite number of discrete lattice and velocity vectors, it can achieve drastic reduction in the number of degrees of freedom (Succi, 2001).

The kinetic nature of the LBM leads to many features; 1) the linear convection operator, similar to that of the Boltzmann kinetic equation, 2) intrinsic parallelism due to nearest neighbor data communications of the movement process and local calculation of the collision process, 3) easy handling of complex boundaries (porous media), and 4) easy incorporation of model interactions essential in simulating complex fluids.

#### 3.3.2 Application towards imbibition in shale

Lattice Boltzmann method (LBM) is an efficient way to simulate flow behavior in channels, and it is suitable to solve problems that involve complicated boundary conditions and multiphase interfaces. Aidun and Clausen (Aidun and Clausen, 2010) summarized the four basic LBM models as the color-fluid model, the pseudopotential model (or Shan-Chen-LBM, SC-LBM), the free-energy model, and the mean-field theory model. The SC-LBM is originated by Shan and Chen (1993, 1994), which is suitable for multiphase flow calculation. Detailed principles, theories, applications and advantages can be found in Shan and Chen (1993, 1994).

LBM has recently been applied in shale. Zheng et al. (2018) built a modified SC-LBM to simulate spontaneous imbibition behavior in a reproduced complicated three-dimensional shale porous structure, with intergranular and organic pores and microfractures included. The pore space is a mix-wetting system because different contents gain different wettabilities. The simulated imbibition process is shown in Fig. 3. From Fig. 3(a) to (f), the imbibition time increases from 0 to  $5 \times 10^4$ . The simulation result shows that at the beginning, large flowing channel is blocked by liquid. As imbibition time increasing, liquid fragment in fractures gradually decreased. And then, flowing channels are reopened again. This simulation result is significant. First, the result supports automatic

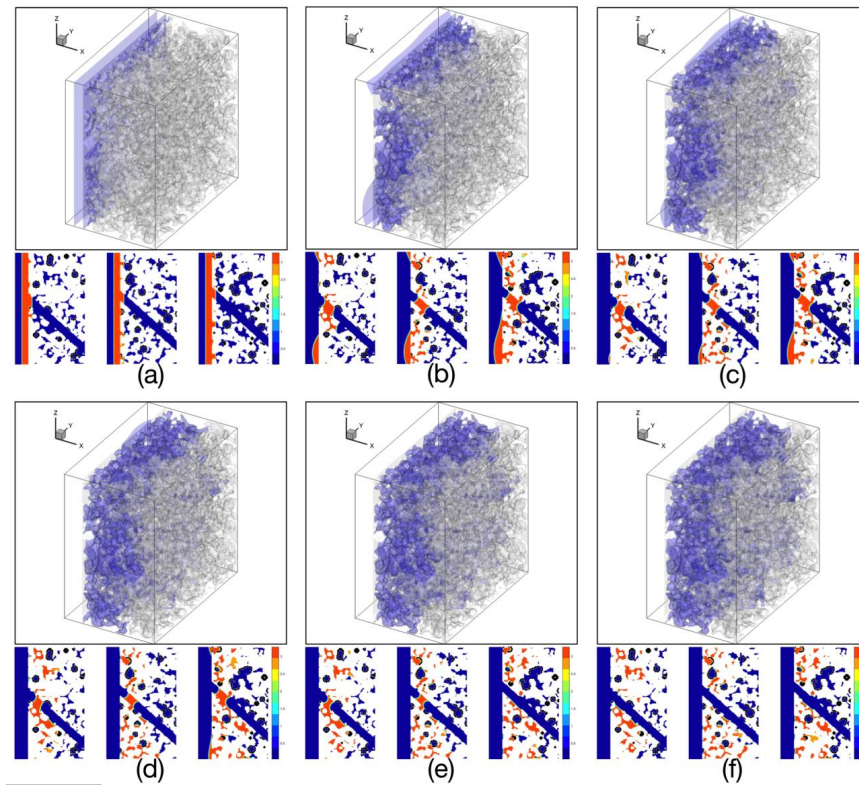
unblock theory for spontaneous imbibition in shale. Second, it provides an explanation for the in-situ observations in which shale gas wells with a low flowback rate could have high production. Third, it explains the EOR mechanism for shut-in operation in shale gas wells. Apart from Zheng et al. (2018), some other studies have investigated imbibition by LBM (Yan et al., 2018).

Apart from Zheng, several other studies have investigated imbibition by LBM. Gu et al. (2019) used improved LBM to calculate counter-current imbibition in pore networks and successfully provided a simulated time-dependent oil recovery factor. The conclusion is that the oil recovery factor is linear with the square root of time for various injection velocities, viscosity ratios, and surface tension. The limitation of this model is its considerable simplicity. It cannot express an occasion when imbibition index  $n$  is not 0.5. Other parameters special in shale need to be considered in future works.

However, several problems must be solved before LBM can be widely used in spontaneous imbibition calculation. The first is viscosity ratio. From our practical experience, the LBM model can hardly obtain iteration convergence when calculating multiphase flow with a large viscosity ratio (especially larger than 10). Thus, using the LBM model for oil-water simulation is acceptable, but additional works should be conducted before it is used on gas-water simulation. The second is calculation complexity (both time complexity and space complexity). For shale with complicated networks, long-term or large-scale imbibition is difficult to compute due to the limitation of computers. Recently, Santos et al. (2018) provided a deep learning method to aid the LBM calculation process. They trained a neural network to understand the relationship between pore-scale morphology and simulation outputs. During training, the neural network would presumably find a spatial correlation at different scales and finally deduce the error. Preliminary results indicated that the deep learning method could reduce the prediction error from 29.24%-40.75% to 5.74%-6.93% at different kernel sizes. Although they calculated pressure and velocity field for one phase in normal grain, it is a promising method to use in the future.

#### 3.4 Capillary bundle and pore network method

The capillary tube model, which was introduced by Lucas (1918) and Washburn (1921), laid the foundation for spontaneous imbibition modeling. Several studies have emerged on the basis of the capillary tube model and its revised models. Many reviews have summarized the development of capillary models (Cai et al., 2001, 2010a, 2010b, 2010c, 2012). In this section, we briefly review the development of the capillary tube and capillary bundle models. Li et al. (2018b) introduced dynamic contact angle and wall influence into an imbibition model for nanopores, used the phase portrait analysis method, and derived solutions in analytical and numerical forms. Shi et al. (2018) extended the previous model by assuming nonpiston-like imbibition in capillaries with different diameters. Mumuni and Pegg (2018) derived a revised capillary bundle model to integrate the fractal dimension of a sample. Wang and Zhao (2019) presented a semianalytical model to



**Fig. 3.** Simulated imbibition process in shale (Zheng et al., 2018). The upper row is spontaneous imbibition simulation results at different time steps (imbibed liquid is blue, pore space is gray). The lower row is the density distribution in the slices of  $Y = 10, 80, \text{ and } 150$ . (a) is initial stage, (b)-(f) is imbibition time at  $1 \times 10^4, 2 \times 10^4, 3 \times 10^4, 4 \times 10^4$  and  $5 \times 10^4$ , respectively.

simulate the cocurrent imbibition process in tight oil reservoirs based on the tortuous capillary tube model and fractal theory. However, the capillary-bundle-originated imbibition model for special structures in shale is uncommonly seen.

The pore network model is another model that simplifies the pore space of porous media. Since the pioneering work of Fatt (1956), the pore network model has been widely used to analyze force and spontaneous imbibition. Fischer et al. (1999) introduced the capillary pressure curve into the pore network model. Øren et al. (1998), Øren and Bakke (2003) and many other studies have developed this model gradually, and this part has been summarized by many works. Wang and Sheng (2018) built a quasistatic pore network model to simulate drainage and imbibition in shale.

### 3.5 Handy's model

The traditional imbibition model originated from Handy and is shown as

$$Q_w = \sqrt{\frac{2P_c k_w \phi A^2 S_w t}{\mu_w}} \quad (9)$$

where  $P_c$  is capillary force,  $k_w$  is intrinsic permeability,  $\phi$  is permeability,  $A$  is the area of cross section,  $S_w$  is normalized water saturation,  $\mu_w$  is viscosity of water. Handy's model is a water-gas system. Based on Eq. (9), Distefano et al. (2017) derived a set of nonlinear second-order differential equations,

and provided their analytical solutions, which are applied to the measured imbibition data to determine effective contact angles. Makhanov et al. (2012) suggested that in Eq. (9), the item  $P_c k_w S_w$  can be regarded as imbibition potential, which is a useful parameter to quantify the imbibition ability of porous media. Hu et al. (2012) and Makhanov et al. (2012, 2014) suggested that these parameters should meet the following equation:

$$Q_w = At^n \quad (10)$$

where  $A$  denotes the imbibition rate, which reflects the speed of imbibition at the initial stage; and  $n$  denotes the imbibition index. According to Handy's model,  $n$  should be 0.5. On the contrary, from an experiment of shale imbibition, as Hu et al. (2012) summarized recent experiments,  $n$  is always lower than 0.5 in shale. Hu et al. (2015a) suggested that the low pore connectivity in shale is responsible for an  $n$  lower than 0.5, whereas Liu et al. (2016a, 2016b) suggested that such low connectivity is due to the strong solid-liquid interaction.

The expression of imbibition rate  $A$  (defined in Eq. (10)) is an important parameter to quantify the spontaneous imbibition of hydraulic fracturing fluids into shale. McWhorter and Sunada (1990) presented the expression as

$$A = \sqrt{\frac{\phi}{2} \int_{S_i}^{S_o} \frac{(S_w - S_i) D(S_w)}{F(S_w)} dS_w} \quad (11)$$

where  $\phi$  denotes the porosity;  $S_i, S_o, S_w$  are initial wetting fluid saturation in matrix, time-invariant wetting fluid saturation at the fracture face and average saturation;  $D$  is akin to a diffusivity and  $F$  is the fractional flow function, defined by McWhorter and Sunada (1990). Birdsell et al. (2015a, 2015b) provided some computations for imbibition in partially saturated shale based on an exact semianalytical solution, and provided the expression of facilitated  $A$  with shale properties by introducing revised capillary pressure-saturation curves (Pc-Sw). The expression of revised  $A$  is:

The expression of revised  $A$  is

$$A(S_i) = \sqrt{\frac{\phi}{2} \frac{k(S_w^x - S_w^f)}{\mu_w \alpha_i}} A^*(\tilde{S}_{ei}) \quad (12)$$

where  $A^*$  is expressed as

$$A^*(\tilde{S}_{ei}) = \sqrt{\int_{\tilde{S}_{ei}}^1 \frac{(\tilde{S}_e - \tilde{S}_{ei}) D^*(\tilde{S}_e, S_e)}{F(\tilde{S}_e)} d\tilde{S}_e} \quad (13)$$

where  $A^*$  is nondimensional imbibition rate parameter;  $k$  is instinct permeability;  $S_w^f$  denotes the saturation attained by any particular scanning imbibition curve as the infinity of capillary pressure,  $S_w^x$  denotes the wetting phase saturation corresponding to nonwetting phase residual saturation, detailed definitions are shown by Gerhard and Kueper (2003).  $S_e$  is effective saturation, defined by van Genuchten (1980);  $\tilde{S}_e$  and  $\tilde{S}_{ei}$  are modified effective saturation and initial modified effective saturation, defined by Birdsell et al. (2015a). In accordance with Birdsell's simulations, in a gas-liquid system, most fluid volumes (15%-95%) can be imbibed into shale during hydraulic fracturing; in an oil-liquid system, the imbibed fluid volume is much lower (3%-27%). However, the model also has limitations wherein the adsorption of water into clay minerals, the effect of microfractures, the direction of imbibition compared with lamination, osmotic potential, or other chemical interactions are ignored (Birdsell et al., 2015a).

Other models have been developed recently. Based on continuum function, Ren et al. (2016) presented a spontaneous imbibition model considering a hysteretic relative permeability-saturation-capillary pressure relation. In this model, the hysteretic formulations, capillary pressure, and relative permeability were influenced by current saturation and the history of saturation in the invaded zone. This study provided a new insight into fracturing fluid imbibition during the shut-in period. Qu et al. (2019) developed a coupled model for water spontaneous imbibition in shale considering the influence of clay swelling and shale damage. Meng et al. (2019a, 2019b) built partially water-covered matrix blocks with different boundary conditions and investigated oil recovery through spontaneous imbibition under different boundary conditions.

Soulaine et al. (2019) built a microcontinuum framework based on an FIB-SEM image from shale and calculated multiphase flow according to the microcontinuum model. However, water invasion of the clay layer by suction, osmotic, and capillary forces was not considered. The imbibition mechanisms should be considered in further investigations. Models

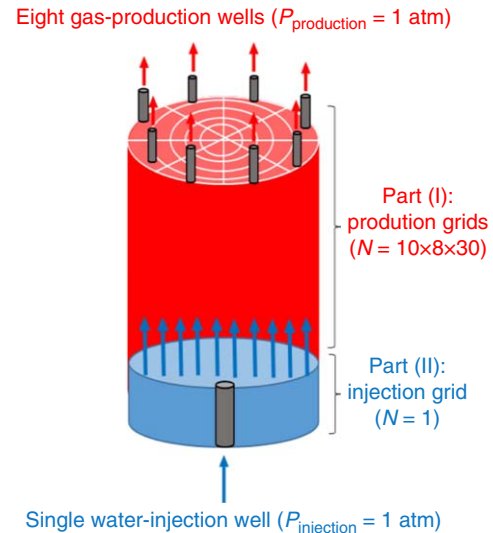
based on governing equations also need discretization to derive numerical solutions. Bridging the large gap between computational efficiency and reliability is a problem. Tchelepi and his fellows developed a multiscale formulation for multiphase flow in complex 2D and 3D grids originating from real samples. This method greatly decreased the calculation time, thereby making multiphase flow in 3D space computable. Related studies can be found in (Guo et al., 2018a, 2018b, 2019; Mehmani and Tchelepi, 2018; Soulaine et al., 2018; Wong et al., 2019). The method can potentially calculate spontaneous imbibition in the future.

### 3.6 Continuum-scale physics simulation method

Apart from modeling approaches discussed above, another method to model imbibition in shale is based on continuum physics implemented in typical reservoir simulators (CMG, ECLIPSE), which solve for mass, momentum, and energy (for heat transport) balance equations at the continuum-scale. Yassin et al. (2016) suggested that shale has dual wettability systems. The organic part is strongly oil wet and prefers to imbibe the oleic phase, whereas inorganic parts are usually hydrophilic and prefer to imbibe aqueous phase. They extended traditional unified wettability into dual wettability systems in which water and oil can act as wetting phases in the organic and inorganic pores, respectively. A co-current imbibition model with dual wettability systems is built to simulate spontaneous imbibition in shale, as shown in Fig. 4.

The model could be successfully solved using a commercial simulator (ECLIPSE 100). Results (Fig. 5) showed that the dual wettability model presented a better match with measured data compared with the uniform wettability model. Hence, dual wettability of organic and inorganic pores is unneglectable for shale imbibition.

Ghanbari and Dehghanpour (2016) used the flowback data



**Fig. 4.** Schematic of simulation model for concurrent imbibition (Yassin et al., 2016). The imbibition simulations were performed using ECLIPSE, a reservoir simulator with continuum-scale physics.



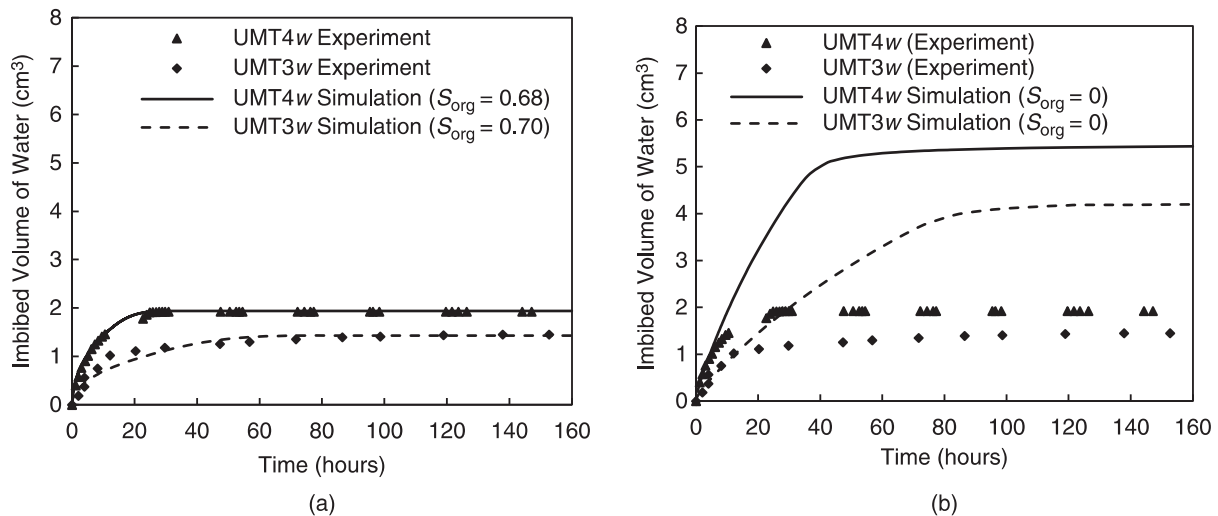


Fig. 5. Comparison between measured and simulated imbibition curve in dual-wettability and uniform wettability models (Yassin et al., 2016).

of 18 shale gas wells to investigate the parameters controlling the fluid production through a commercial simulator (CMG) for post shut-in period when the fracturing fluid imbibes into the shale. Their study concluded that the counter-current imbibition of fracturing water (water imbibing into the matrix and pushing gas out towards the fractures) can lead to gas build-up in the fractures that increases the early-time gas production rate. Similar outcome was hypothesized by Singh (2016) in their review based on the interpretation of the post shut-in period production data.

#### 4. Factors influencing spontaneous imbibition in shale

Shale plays contain abundant hydrocarbons, where most of these are stored in micro/nanopores in matrix and microfractures. Many important factors influencing imbibition, such as pore structure, pore size distribution, wettability, types of flowing space (organic pores/inorganic pores/ micro slits), connectivity, capillary force, mineral component, liquid flowing behavior, etc., have been the focus of recent studies on spontaneous imbibition in shale, where some of these factors are reviewed below.

Possibly the most important factor influencing spontaneous imbibition in shale are the type of pores in shale. The primary space for hydrocarbon flow and storage in shale plays is within organic/inorganic pores, microslits, and fractures. In terms of the pore volume, majority of the pore space in shale consists of organic and inorganic micropores. Inorganic pores commonly consist of clay, interparticle, and intragranular pores, especially clay pores (Loucks 2009, 2012) some of which may swell after interacting with fracturing liquid. Roughness on the pore wall is another influencing factor which should be considered (Supple and Quirke, 2004, 2005). Singh (2016) reviewed the impact of type of pores and the pore size distribution on spontaneous imbibition in shale and how it impacts the shape of the imbibition curve in terms of the initial and late time curve exponents.

The second factor is wettability and the variety of pores that can greatly influence capillary force and, as a result, imbibition behavior. From the view of wetting systems, micro-wetting characteristic is a mixed and complicated system that also depends on the radius of the droplet (Singh, 2016). Organic pores are generally oil wetting, whereas inorganic pores can be oil wet, water wet, and mix wet, depending on the minerals and roughness on the pore wall. Wettability is influenced by contact surface and liquid behavior, which have been shown to influence electrical properties, capillary pressure, waterflood behavior, relative permeability, dispersion, and simulated EOR (Anderson, 1986). Wettability is possibly one of the most popular characteristic of a rock that attracts considerable attention in shale imbibition analysis, possibly because it is a well-known topic in petroleum engineering, and unlike other measurements (core flooding) it does not require an extensive experimental setup. Wettability has been studied for different applications in petroleum engineering, where one of the most well-known application is for chemical-enhanced oil recovery in which surfactant is used as an agent to change the wettability of the oil-wet rock to water-wet that helps displace oil out from smaller pores. In altered water-wet formation, additional water can be imbibed into the oil-containing matrix spontaneously, thereby driving substantial oil out of the matrix. This method has been widely studied and has been successfully used in sandstone, chalk, and carbonates in recent decades (Standens and Austad, 2000; Høgnesen et al., 2004; Hirasaki and Zhang, 2004; Strand et al., 2008; Sheng, 2013), but seldom in shale. Sheng (2017) investigated the influence of wettability modification on shale production by using numerical simulation. Findings showed that using surfactants in shale to achieve ultralow interfacial tension may be ineffective. Singh and Cai (2018) used numerical simulations to investigate enhancing oil recovery from shale formation through surfactant injection that reduces the interfacial tension between oil and water, which also concluded that reducing interfacial tension of fluids in shale is not as effective in

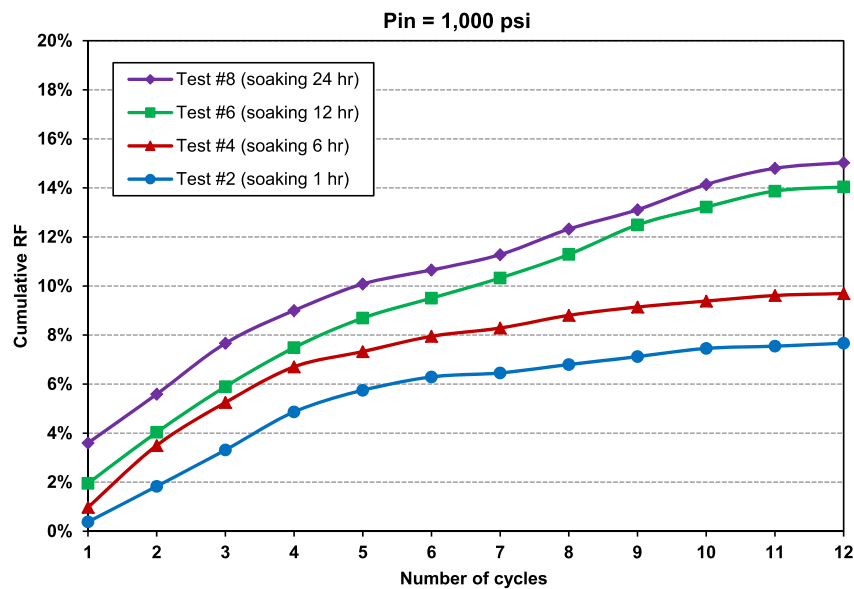


Fig. 6. Oil recovery performance of the water huff-n-puff with different soaking times (Yu et al., 2017).

driving the oil out as in conventional oil recovery. Related works have been investigated by Shuler et al. (2011), Wang et al. (2011) and Xu et al. (2015). Another method has been studied to alter liquid behavior, including salinity, acidity, and alkalinity, to enhance the potential of water imbibition in shale plays and hydrocarbon recovery (Morsy et al., 2013a, 2013b).

The third factor, although not very popular in the literature, is osmotic pressure. The shale saturated by high salt concentration sucks liquid from shale saturated by low salt concentration through a semi-permeable membrane (clay) spontaneously (Singh, 2016), which leads to existence of osmotic pressure that can be as large as about 30 MPa for formations with about 10% porosities and less for higher porosity medium (Singh, 2016). Singh (2016) provided an extensive review on the effect of osmosis in shale and how it may potentially impact imbibition behavior. Osmosis is triggered due to chemical potential existing between brines of different salinities in the formation, which in general is not observed at laboratory-scale experiments where either pure water or brine of uniform salinity is used. Also, as discussed in Singh (2016), the osmotic pressure developed in the laboratory using brines of different salinities may not as high as the estimates in the field due reasons such as lower efficiency of membranes, lower specific area, etc. However, there are no major studies on this topic besides the work by Fakcharoenphol et al. (2014) who found that osmotic pressure can push the oil out from fractured shale formations through counter-imbibition.

Microcracks can greatly influence imbibition. Experiments show that shale samples with imbibition direction parallel to its bedding plane gain much higher imbibition speed compared with samples with imbibition direction perpendicular to its bedding plane (Ghanbari and Dehghanpour, 2015; Xu et al., 2019). Wang et al. (2011) observed from oil-water counter-current imbibition that in a shale matrix, some microcracks began to emerge, and the cracks' direction was parallel to

the shale's bedding plane. These cracks could greatly increase the imbibition ability of shale samples. Liu et al. (2019) found that the stress field can greatly influence the number of cracks. Singh (2016) reviewed the creation of microcracks, its impact on imbibition, and the criterion for initiation of micro-fractures due to imbibition in shale.

In addition to the influencing factors discussed above, clay hydration (Singh, 2016) is another factor that can imbibe water in shale. Specifically, clay hydration is a process through which clays inside the shale matrix can absorb water through development of an electrostatic double layer (EDL) on the pore wall. Per the EDL concept in shale (Singh, 2016), clay surface that is negatively charged is associated with two layers of positive charges with varying thicknesses derived from the bulk aqueous solution. Binazadeh et al. (2016) found that in unwashed shale powder, deionized water is imbibed faster than brine. It is believed that the imbibition rate depends on the thickness of the electrostatic double layer (Singh, 2016; Binazadeh et al., 2016).

There are several ways by which deeper understanding of spontaneous imbibition can be used to engineer a better oil/gas recovery in the field, such as extending well shut-in time, increasing the number of water huff-n-puff cycle, etc. During the shut-in period, wetting fracturing liquid can be imbibed into shale matrix, replace oil and gas, and enhance recovery. Several shale gas wells have designed the shut-in period before gas production to increase the initial production rate (Abbasi et al., 2014; Bertoncello et al., 2014). A longer shut-in period for water-wet shale has shown to reduce the water production rate, in some cases almost no water is produced after shut-in, and increase oil/gas production rate (Singh, 2016). Yu et al. (2017) examined the effects of soaking time and injection pressure on the recovery performance based on cyclic water injection (water huff-n-puff) experiments, as shown in Fig. 6. Within a certain range, a long soaking period caused a high

recovery in the water huff-n-puff process. Once the recovery of the system approached maximum recovery, extending the soaking period would no longer increase recovery.

## 5. Potential ideas for future research

This work reviewed recent advances in spontaneous imbibition in shale, including modeling and experimental techniques to study spontaneous imbibition in shale. Here, we present the discussions and expectations in three parts, namely, mathematical models, experiments, and the mechanisms that need to be focused on.

From perspective of mathematical modeling, most theoretical models are still based on classical imbibition models, including several revisions from capillary bundle, PNM, and Handy's models. Pore space for shale is much more complicated than those of conventional rocks such as sandstone. The connectivity, mixed wettability system, and non-Darcy effects in nanopores should be carefully focused on in these models. Combining classical models with real images acquired by SEM, FIB, EDS, CT, etc. has potential. For example, basic data such as the wettability for organic and inorganic pores in a volume larger than REV (Singh, 2014, 2017) can be extracted first, and then the classical models can be built on the basis of this data specific to shale, such as extending the capillary bundle model into two or more wettability systems. The revised models that account for realistic pore and wettability properties of shale acquired through imaging techniques may be more suited to investigate imbibition in shale. An example of this approach is used in predicting permeability of shale by identifying its features through acquired images (Singh and Cai, 2019a, 2019b). For the MD and LBM simulations, a major drawback is computational complexity along with limitations with spatial and time scale that can be simulated. It is almost impossible to scale-up MD and LBM simulations from pore-scale to field-scale estimates without employing continuum-scale models. However, MD and LBM methods have potential to investigate relative strength of various processes (capillarity, osmotic pressure, clay hydration/EDL, etc.) driving spontaneous imbibition at the pore-scale. These two methods (MD and LBM) can also benefit greatly with the application of deep learning, as shown in studies by Santos et al. (2018) and Guo et al. (2018b). Combining deep learning with MD or LBM method may greatly accelerate their computational prediction speed, making it possible to simulate multiphase imbibition directly on complicated 3D pore space on a lab scale (pore diameter 2.5 cm) for several days.

From experimental perspective, balance weight remains a dominant and economic method for shale imbibition tests. To the best of our knowledge, the imbibition curve for one shale rock may not be repeatable for another rock, even possibly from within the same reservoir. This is because there are many different variables that affect imbibition in shale, including the swelling of clay minerals, generation of microcracks, diffusion of ion, instability of core, etc., all of which make the imbibition process in shale non-uniform and complex. For scanning instruments, maintaining the increase in resolution is unrealistic and uneconomic. Each method has its own best and

economic resolution. Confirming a REV and then combining two or more test results by using several mathematical algorithms in this REV are necessary and promising. For example, cross-correlation-based simulation (CCSIM) is an encouraging method to combine image information on different scales, such as acquiring organic pore information from FIB, inorganic pore information from SEM and slit information from micro-CT (Tahmasebi, 2015, 2016; Tahmasebi and Sahimi, 2016).

Imbibition behavior in shale is a complicated process that is affected by several variables, such as clays, brine salinity, microcracks, wettability, types and sizes of pores, etc. The models and experimental techniques mentioned above have limited capability in capturing or investigating few factors out of all the factors that drive imbibition, therefore, these techniques should be carefully chosen based on the focus variable and one should also be aware of their limitations. Several years ago, high capillary pressure in shale hypothesized by presence of nanometer-sized pores was considered to be an important research area for spontaneous imbibition in shale. With recent developments, ion diffusion, osmotic pressure, mixed-wetting and hemi-wicking have attracted increasing attention in shale imbibition research, which is an enlightening trend. The mystery of spontaneous imbibition in shale, especially the key factors deriving the spontaneous imbibition process and applying that understanding to enhance shale oil/gas production would gradually be uncovered with the development of models, experiments, and mechanisms.

## 6. Conclusions

This study reviews the recent advances in spontaneous imbibition in shale, including recent theoretical models, experiments, and mechanisms. Although these advances have led to increase in our understanding of imbibition in shale, it also reinforces the complexity associated with the spontaneous imbibition such that specific questions on several fronts require more clarity as discussed under previous section. It is evident from the literature that we are far from completely understanding the imbibition process in shale, especially because there is not a single study that tries to uncover the individual strength of myriad number of variables driving the spontaneous imbibition in shale. This combined with non-repeatable pattern of imbibition curves even for rocks from the same formation are incomprehensible with our current knowledge and understanding.

Several theoretical models are available for explaining spontaneous imbibition, including co-current and counter-current imbibition. These models remain useful for shale imbibition analysis under specific conditions through continuous revisions and development. For analytical models, including capillary bundle model, pore network mode, and continuum model, there is a scope is to introduce characteristic shale properties into models. For micro and pore-scale numerical models, the key is to scale-up MD and LBM simulations from pore-scale to field-scale estimates along with overcoming the obstacle of computational time.

On experiments, apart from balance-weighting experiments, several new detecting methods and instruments have

been gradually used in spontaneous imbibition observations in shale, especially the collaboration of several experiments, including NMR, CT, and balance weighting, which have extended the capability to probe imbibition and provided additional ways to evaluate the imbibition behavior in shale. Many key factors impacting spontaneous imbibition in shale were considered and applying that understanding to enhance shale gas/oil production was also discussed.

From the perspective of mechanisms, studies have gradually changed from focusing on high capillary pressure hypothesized by presence of nanometer-sized pores and strong wetting contact angles to focusing on other factors, such as ion diffusion, osmotic pressure, mixed wetting, and hemiwicking theory. With the further development of spontaneous imbibition research, the mystery of mechanisms driving spontaneous imbibition in shale would be gradually uncovered in the future.

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### Conflict of interest

The authors declare no competing interest.

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