

## Original article

## A model of spontaneous flow driven by capillary pressure in nanoporous media

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doi:10.26804/capi.2020.01.01.**Abstract:**

The spontaneous capillary imbibition phenomenon is a fundamental mechanism in porous media with applications in many fields. In low permeability shale reservoirs, this flow driven by capillary pressure is extremely important due to the predominance of nano-scale pores, which enhance capillary pressure and weaken hydrodynamic viscous force. This paper presents the results of an analytical model for capillary rise in nano-channels by taking into consideration the effect of inherent surface roughness. Model predictions match better with available experiments results. Relevant experiments were carried out on silicon-based nano-channels with rectangular section, which height is between 5 and 50 nm using de-ionized water. Results proved that the capillary rise kinetics in nano-channels follows the modified Lucas-Washburn law. The surface roughness adds extra resistance during the process of capillary rise, which is calculated as an equivalent porous medium layer. The capillary model is extended to porous media using the capillary bundle concept. In this model, imbibition height versus time was defined. Using this equation, the weight of imbibed liquid by spontaneous imbibition can be obtained. The results from this study demonstrate that the spontaneous imbibition in nanoporous media could be scaled and predicted.

**1. Introduction**

Along with the expanding unconventional development, the spontaneous imbibition mechanism turned out to be a hot topic in petroleum engineering. The hydrodynamic flooding is limited due to the nano-pores and nano-channels, which are widely developed in formations. Spontaneous imbibition is an effective way to drive the oil out of nanoporous matrix media. Thus, it is essential to understand the dynamic characters of spontaneous imbibition in these media with nano-pores. A great interest has been focused on this topic for more than a century, with many models developed (Handy, 1960; Morrow and Mason, 2001; Alava et al., 2004; Schmid et al., 2016; Pavuluri et al., 2018; Abd et al., 2019).

The capillary rise process can be divided into two regimes: short-time and long-time. At the beginning of fluid invasion, the gravity can be ignored, since the intake fluid weight is limited and inertia force dominates this rise process (Quére, 1997, 2008; Dhar et al., 2016). As the imbibition height increases, viscous force becomes the main flow resistance and the effect of inertia force decreases. During this regime,

the most popular model for capillary rise is called Lucas-Washburn (LW) equation, which was proposed by Lucas in 1918 and improved by Washburn in 1921. The model gives an analytic solution for the height of capillary rise, based on the hypothesis of capillary bundle model (Lucas, 1918; Washburn, 1921). Afterwards, most of the capillary rise models were developed base on LW equation. Various parameters were taken into consideration, some theoretical models considered the gravity (Tavassoli et al., 1991) while others considered the pore geometrical shape (Hammecker et al., 2011; Cai et al., 2014). There are also other parameters like the tortuosity of capillary and fractal dimensions of porous media (Cai et al., 2010; Cai and Yu, 2011), the interfacial tension (IFT) of liquid system (Schechter et al., 2005) and inertia force (Zhmud et al., 2000). Mattax and Kyte (1962) first presented dimensionless time  $t_D$ , and gave the relationship between recovery and time, which was Mattax-Kyte (MK) model. For a more accurate result, the shape factor was introduced into this model by Kazemi et al. (1992). Zhang et al. (2008) carried out a set of lab experiments with four different boundary conditions. The results also verified Kazemi's model with shape factor.

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Li and Horne (2001, 2004) carried out laboratory imbibition experiments with water and gas, using circular tubes filled with glass beads. They put forward an imbibition model with phase saturation, and modified the model later by considering wettability, dynamic capillary force as well as gravity for oil and water phases. An effective viscosity term which considered the ratio of wetting and non-wetting phase, was proposed on counter-current spontaneous imbibition by Mason et al. (2010), and the model got a better match with imbibition experiments even with various boundary conditions. Wang and Shen (2018) introduced capillary fractional flow function into Buckley and Leverett theory and proposed an self-similar analytical solution for imbibition in porous media. Meng and Cai (2018) demonstrated solutions for spontaneous imbibition with various boundary conditions. Fries and Dreyer (2008) proposed an analytical solution for capillary rise considering gravity. The roughness of capillary was first mentioned in the capillary rise process, which might be the reason for a lower theoretical evaluation.

Surface effects become more significant when the flow scale decreases. Surface roughness is the most common factor in microscale surface effects (Mala and Li, 1999). In ultra-low permeability reservoir, the relative surface roughness could reach up to 25%, which represents a high level of resistance coefficient, and results in the non-negligible of surface roughness (Du et al., 2013). There are mainly 4 methods for describing surface roughness, the fractal geometry method (Gamrat et al., 2009), the porous medium layer (PML) method (Koo and Kleinstreuer, 2005), the roughness-viscosity model (Mala and Li, 1999) and the regular perturbation method (Van Dyke, 1975).

Our studies are based on the imbibition phenomenon with wall-roughness in nanoporous media. We calculated the frictional resistance by assuming inherent obstacles on the wall surface. This represents a layer of porous media on the wall surface, so the extra flow resistance caused by roughness in the capillary rise process is equal to the flow resistance in porous media. The purpose of this paper is to analyze the capillary rise considering wall roughness. As a result, an analytical solution for a nano-scale capillary rise is obtained.

## 2. Resistance of equivalent roughness PML

From many nano-channel experimental results, we can see that the imbibition height is always lower than what LW equation expected, although the driven force of capillary rise is enhanced due to the nano-diameter of capillary, indicating that there are more resistances should be taken into account during the rising in nano-capillary. For the purpose of better matching and understanding of the mechanics, we built a model for spontaneous imbibition in single capillary with wall roughness, which obtains a better consistency with the results in experiments. This model is based on Newton's second law, which gives an analytical solution as shown in the previous paper (Shen et al., 2017). Here is a simple introduction of this single capillary spontaneous imbibition model in nano-channels.

The kinetics for a viscous, non-compressible liquid rise

in a vertical cylindrical nano-channel with inherent surface roughness is

$$F_c - F_{vis} - F_g - F_r = ma \quad (1)$$

The lefthand side terms in Eq. (1) is the capillary force  $F_c$ , viscous force  $F_{vis}$ , gravity force  $F_g$  and frictional resistance  $F_r$  due to wall roughness. The right side is inertia force, which  $m$  refers to mass imbibed into channel and  $a$  refers to accelerated velocity of rising meniscus. The effect of gravity on nano-capillary was discussed before, the capillary rise height results of LW equation with and without gravity are identical (Shen et al., 2017). Since the diameter of capillary here is quite small, the gravity is ignored.

Based on the Young-Laplace equation, the capillary pressure  $p_c$  for a cylindrical capillary is

$$p_c = \frac{2\sigma \cos \theta}{R} \quad (2)$$

where  $\sigma$  is the surface tension of liquid-gas interface,  $\theta$  is the contact angle, and  $R$  is the pore radius. So, Eq. (1) can also be written as

$$\frac{2\sigma \cos \theta}{R} - \frac{8\mu}{R^2} \frac{dh}{dt} h - \Delta p = \rho \frac{dh'}{dt} \quad (3)$$

where  $\mu$  is the liquid viscosity,  $h$  is the height of capillary rise,  $\rho$  is the density of the fluid, and  $\Delta p$  is the pressure loss due to wall roughness. For the calculation of resistance force, the method of PML is used. We assume that there are obstacles on the wall, with the shape of cylinders. The height and radius are  $\varepsilon$  and  $r$  respectively and the distance between two adjacent cylinders is  $l$ , as shown in Fig. 1. The resistance force of the wall is calculated as the fluid flow resistance in the cylinders forest.

The flow resistance of the PML  $\Delta p$  is composed of frictional drag  $F_f$  caused by viscous friction, and the local resistance  $F_D$  caused by flow around the cylinder, which can be expressed as (Spurk and Aksel, 2008)

$$\Delta p = F_f + F_D \quad (4)$$

where

$$F_f = \lambda \frac{u^2 \rho}{2} \frac{S}{V}$$

$$F_D = \frac{1}{2} C_D \rho u^2 N$$

where  $\lambda$  refers to the friction drag coefficient,  $u$  is the average flow velocity,  $C_D$  is the coefficient of local resistance, and  $N$  is the projected area perpendicular to the direction of flow motion. Considering one micro-unit is made up of four cylinders,  $S$  refers to the surface area of one micro-unit, and  $V$  is the volume of micro-unit.

$$S = 2(l^2 - \pi r^2) + 2\varepsilon l(l - 2r) + 2\pi r \varepsilon \quad (5)$$

$$V = \varepsilon l^2 \quad (6)$$

Due to the low flow rate in capillary rise, the Reynolds number  $Re$  is low, so the friction coefficient  $\lambda$  for laminar flow is (Spruk and Aksel, 2008)

$$\lambda = \frac{64}{Re} \quad (7)$$

and the Reynolds number  $Re$  is calculated by

$$Re = \frac{\rho u d_e}{\mu} \quad (8)$$

where  $d_e$  is the characteristic length. Since the irregular section of PML, the characteristic length  $d_e$  of a non-circular tube is defined as four times the hydraulic radius  $r_e$ .  $r_e$  is calculated as the effective section divided by the wetting perimeter, so  $d_e$  is

$$d_e = 4r_e = \frac{4V\phi_{PML}}{S} \quad (9)$$

where  $\phi_{PML}$  refers to the porosity of PML, and it is calculated by

$$\phi_{PML} = \frac{\varepsilon(l^2 - \pi r^2)}{\varepsilon l^2} = 1 - \pi\left(\frac{r}{l}\right)^2 \quad (10)$$

Substituting Eqs. (5), (6) and (10) into Eq. (9), we can get  $d_e$  as

$$d_e = \frac{2\varepsilon(l^2 - \pi r^2)}{l^2 - \pi r^2 + \varepsilon(l - 2r) + \pi r\varepsilon} \quad (11)$$

The average flow velocity  $u$  and Darcy velocity  $v$  in the PML are related via

$$v = u\phi_{PML} \quad (12)$$

Thus,  $F_f$  is obtained

$$F_f = \lambda \frac{u^2 \rho}{2} \frac{S}{V} = \frac{32\mu u [l^2 - \pi r^2 + \varepsilon(l - 2r) + \pi r\varepsilon]^2}{\varepsilon^2 l^2 (l^2 - \pi r^2)} \quad (13)$$

Then in order to calculate the  $F_D$ , we should know about  $C_D$ . The local resistance coefficient  $C_D$  is related to the velocity

distribution. The relationship between  $C_D$  and the frontal drag coefficient  $C_x$  is

$$C_D = C_x \frac{S_m}{N} \left( \frac{u_{mect}}{u_m} \right)^3 \quad (14)$$

where  $S_m$  refers to the sectional area of the cylinder,  $u_{mect}$  is the local velocity, and  $u_m$  is the maximum velocity.

The velocity distribution is nonuniform along the section in the tube, thus the resistance of the object depends on the object position. The frontal drag coefficient  $C_x$  is closely related to  $Re$ . Here  $Re \ll 1$ . The fluid flow around cylinders from the front to the back is smooth, and the inertia force can be neglected. The  $Re$  for nano-capillary rise is ultra-low, therefore, the quadratic term for the velocity can be neglected. So the flow resistance for PML is

$$\Delta p = \frac{32}{d_e^2 \phi_{PML}} \mu v = \frac{32\mu}{d_e^2} u \quad (15)$$

### 3. Model validation and predictions

#### 3.1 Capillary rise restrained by surface roughness

Capillary filling experiments of nano-channels have been carried out many times. Tas et al. (2004) regarded that deviation between LW equation and experiment results was due to the approximately 40% higher of the apparent viscosity in nano capillaries, while Haneveld et al. (2008) believed that the apparent viscosity was elevated by the layers close to the channel walls. The model that considered inherent surface roughness was compared with the experimental results from Haneveld et al. (2008), and the viscosity and surface tension of water were taken from the CRC handbook (Lide, 2001). The width of the rectangular tube  $d$  is 20  $\mu\text{m}$ , the surface tension and viscosity at 20°C are 72.75 mN/m and 1.002 mPa·s, the capillary wall is completely wetted by water, thus the contact angle is 0°. Nanochannels (20  $\mu\text{m}$  wide, 1 cm long) were defined by photolithography and 1% hydrofluoric (HF) etching of the silicon oxide spacer layer, and the filling of the 11 and 5 nm channels was observed using differential interference contrast (DIC), based on AFM image analysis (Fig. 2 in

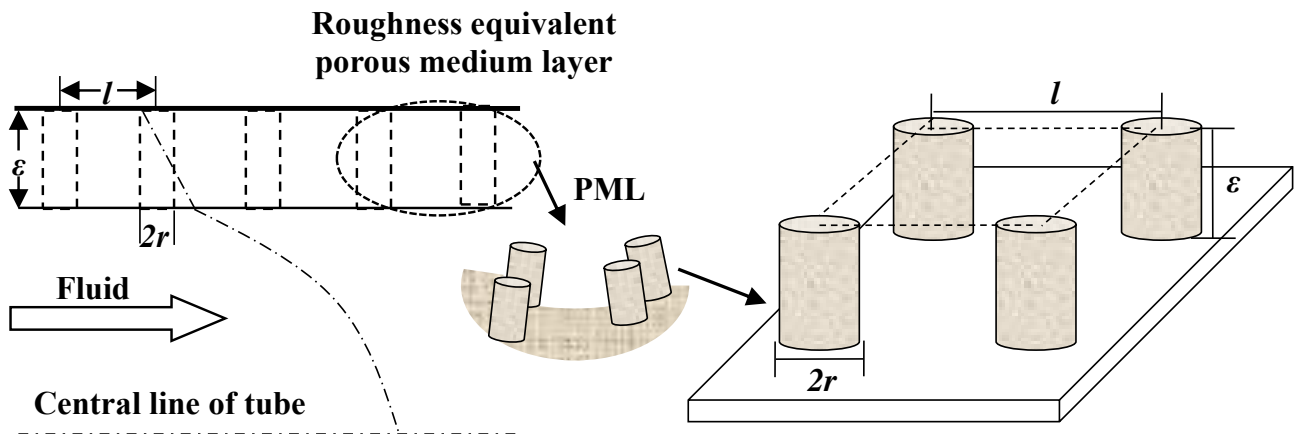


Fig. 1. Schematic diagram of an equivalent roughness PML micro-unit.

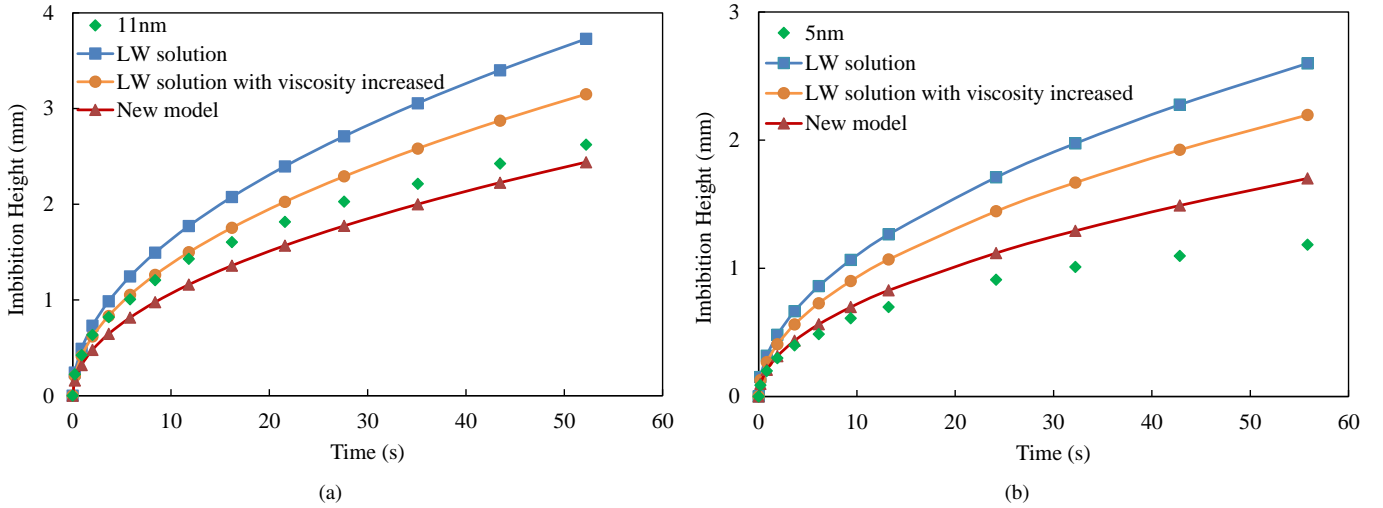


Fig. 2. Different solutions compared with experimental results for nano-capillary filling.

Haneveld et al., 2008), the obstacles on the wall were ignored. In the analytical model, we set the same roughness parameters in these two tubes, where  $l$  is 5 nm and  $r$  is 2.8 nm, which leads to a different surface roughness, and may cause the discrepancy between new model curve and experimental curve. However, the capillary diameter of a couple of nanometers with an asperity height of 0.5 nm should not be seen as a smooth surface (Haneveld et al., 2008). Therefore, the criterion for a smooth nano-capillary is worth to be discussed in future studies. Based on the previous section and LW equation, we obtain the momentum equation for the nano tube imbibition in circular pipe, taking into account its wall-roughness:

$$\frac{2\sigma \cos \theta}{R} = \left( \frac{8\mu}{R^2} + \frac{32\mu}{d_e^2} \right) \frac{dh}{dt} \quad (16)$$

The differential equation, Eq. (16), can be solved for the initial condition  $h(0)=0$ .

$$h = \sqrt{\frac{\sigma t \cos \theta}{R \left( \frac{2\mu}{R^2} + \frac{8\mu}{d_e^2} \right)}} \quad (17)$$

Based on LW equation, the capillary rise height in rectangular tube is

$$h = \sqrt{\frac{\sigma w \cos \theta}{3\mu} t} \quad (18)$$

For a rectangular cross-section of channels, according to the conclusion of previous literatures (Tas et al., 2004; Delft et al., 2007; Haneveld et al., 2008), Eq. (18) could be modified into

$$h = \sqrt{\frac{\sigma t \cos \theta}{w \left( \frac{6\mu}{w^2} + \frac{16\mu}{d_e^2} \right)}} \quad (19)$$

where  $w$  is the depth of the rectangular nano-channel.

$$d_e = 4r_e = \frac{4wd}{2w+d} \quad (20)$$

Table 1. Parameters of filling experiments in Haneveld et al. (2008) and model calculation.

No.	Channel depth $w$ (nm)	Characteristic length $d_e$ (nm)
1	11	43.9
2	5	19.9

where  $d$  refers to the width of the tube. Both  $w$  and  $d_e$  are shown in Table 1.

### 3.2 Fitting experimental data of nanoporous media

To verify the theoretical results, the capillary filling experiments in nano-capillary done by MESA+ Research Institute (Haneveld et al., 2008) can be used as a comparison. The predicted results were given in Fig. 2. Based on the above parameters, the line with diamonds refers to the experimental results in Haneveld et al. (2008), the square points refer to the LW equation solution (Eq. (16)), the circular points refer to the LW equation solution with 40% elevated viscosity, and the triangle points refer to the model solution (Eq. (17)).

The analytic solution of model considering surface roughness has a better match with experimental data. From Fig. 2 we can see that, the LW equation solutions are usually higher than the experimental results for less friction in the smaller size tube like 11 nm and 5 nm, which the friction would be much higher than the bigger size tube. The apparent viscosity elevated solution shows a lower expected of resistance. Since the flow resistance increases as the decreasing of tube size, the apparent viscosity elevated solution by 40% could not be used as an general solution for different size of tubes. The increased viscosity percentage should change as the scale of capillary diameter changes. The model solution considering friction force of surface roughness shows a better match in different tubes, and it proves that the friction caused by wall

roughness should be taken into account in nano structured capillary imbibition.

#### 4. Model developed for spontaneous imbibition in nanoporous media

Base on capillary bundle model (Lucas, 1918), an analytical model of spontaneous imbibition in nanoporous media is developed. It is here assumed that a porous medium (section area  $A$ , height  $L$ ) is consisted of numbers of capillaries with radius  $r_i$ , permeability  $k$ , and porosity  $\phi$ , which is saturated with oil. The height of imbibed water is  $h$ , then the capillary force is

$$F_{pc} = 2\pi\sigma \cos \theta \sum_{i=1}^n r_i \quad (21)$$

Assuming that the distribution function of capillary radius  $r_i$  is  $f(r)$ , so

$$\sum_{i=1}^n r_i \approx n \int_0^{\infty} r f(r) dr \quad (22)$$

Here assuming the term  $E(r) = \int_0^{\infty} r f(r) dr$  is the expected value for capillary radius, so the porosity  $\phi$  equals to  $n\pi E^2(r)/A$ , then Eq. (21) can be written as

$$F_{pc} = \frac{2\sigma\phi A \cos \theta}{E(r)} \quad (23)$$

The forces on the meniscus includes the gravity of imbibed water  $\rho_w g h \phi A$ , the viscous resistance and the frictional resistance due to the wall roughness. The viscous resistance of water is

$$F_{vis-w} = \frac{\mu_w \phi A}{kk_{rw}} (S_w - S_{iw}) h \phi \frac{dh}{dt} \quad (24)$$

where  $S_w$  is water saturation in the two-phase flow region,  $S_{iw}$  is the initial water saturation,  $k_{rw}$  is the relative permeability of water,  $h$  is the capillary rise height. Similarly, gravity of oil on the meniscus is  $\rho_o g (L - h) \phi A$ , and the viscous resistance of oil is

$$F_{vis-o} = \frac{\mu_o \phi A}{kk_{ro}} (L - h) \phi \frac{dh}{dt} \quad (25)$$

The flow resistance of PML for water and oil are

$$\Delta p_{w/o} = \frac{32}{d_e^2} \mu_{w/o} \frac{dh}{dt} \quad (26)$$

So the capillary bundle model can be written as

$$\begin{aligned} & \frac{2\sigma \cos \theta}{E(r)} - (\rho_w - \rho_o)gh - \rho_o gL - \frac{32}{d_e^2} (\mu_w + \mu_o) \frac{dh}{dt} \\ & = \frac{\mu_w h}{kk_{rw}} \phi (S_w - S_{iw}) \frac{dh}{dt} + \frac{\mu_o (L - h)}{kk_{ro}} \phi \frac{dh}{dt} \end{aligned} \quad (27)$$

When the rising velocity is 0, the imbibing height reaches the highest point  $h_{\max}$ , the  $h_{\max}$  is

$$h_{\max} = \frac{2\sigma \cos \theta}{E(r)(\rho_w - \rho_o)g} - \frac{\rho_o L}{(\rho_w - \rho_o)} \quad (28)$$

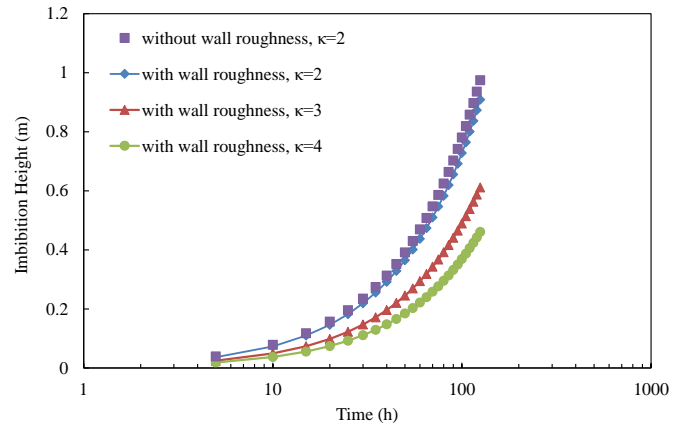


Fig. 3. The position of imbibition front in porous media with different viscosity ratio of oil to water.

Dividing Eq. (27) by  $(\rho_w - \rho_o)g$ , and substituting Eq. (28) into Eq. (27) yield.

$$Bdt = \frac{C + Mh}{h_{\max} - h} dh \quad (29)$$

where

$$M = \phi [\mu_w k_{ro} (S_w - S_{iw}) - \mu_o k_{rw}] \quad (30)$$

$$B = kk_{rw} k_{ro} (\rho_o - \rho_w) g \quad (31)$$

$$C = \phi k_{rw} \mu_o L + \frac{32kk_{rw}k_{ro}(\mu_w + \mu_o)}{d_e} \quad (32)$$

When  $\mu_w k_{ro} (S_w - S_{iw}) - \mu_o k_{rw} = 0$ , the Eq. (29) turns out to be

$$Bdt = \frac{C}{h_{\max} - h} dh \quad (33)$$

So the  $h$  can be solved as

$$h = h_{\max} \left[ 1 - \exp\left(\frac{B}{C}t\right) \right] \quad (34)$$

The equation shares the same form with the one in Fig. 2 with modified coefficients. Based on the height of rise, the imbibition volume can be obtained by  $Q(t) = \phi Ah S_w$ . We assumed that the height of cylindrical core is 10 cm with a radius of 2.5 cm,  $\rho_w = 1000 \text{ kg/m}^3$ ,  $\rho_o = 800 \text{ kg/m}^3$ ,  $\theta = 30^\circ$ ,  $\sigma = 15 \text{ mN/m}$ ,  $\mu_w = 6.5 \times 10^{-4} \text{ Pa}\cdot\text{s}$ ,  $E(r) = 140 \text{ nm}$ , the number of capillaries  $n = 10^{10}$ ,  $k_{rw} = k_{ro} = 1$ , then we calculated that  $\phi = 0.125$ ,  $k = 0.307 \times 10^{-3} \text{ }\mu\text{m}^2$ . For the wall roughness part, the parameters are  $l = 5 \text{ nm}$ ,  $r = 2.8 \text{ nm}$ . Defined that viscosity ratio  $\kappa = \mu_o / \mu_w$ , the effects of wall roughness and viscosity ratio on rise dynamics for a vertical homogeneous porous medium are shown in Fig. 3. As we can see, with the same viscosity ratio, due to the additional resistance which comes from the wall roughness, the rise height of nano-pores media is lower, comparing with the one without wall roughness. The rising height also decreases as the viscosity ratio goes up.

## 5. Conclusions

An analytical expression for calculating the capillary imbibition in nanoporous media has been derived, which considers the inherent wall roughness. The resistance induced by wall roughness is calculated as the resistance of porous medium layer on the wall. With the additional friction force, the capillary rise stops at a lower level comparing with the regular capillary rise height. By means of capillary bundle model, the imbibition model in porous media is also obtained. We can also conclude that as the viscosity ratio of oil to water goes up, the imbibition height goes down, which means that reduce the viscosity ratio of oil to water is also helpful for imbibition recovery.

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## Conflict of interest

The authors declare no competing interest.

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