



## Full Length Article

## Effects of low-salinity waterflooding on capillary pressure hysteresis

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## HIGHLIGHTS

- Connection between interfacial viscoelasticity and capillary hysteresis was further established.
- A hybrid dynamic-quasi-static capillary pressure method was tested for low-salinity waterflooding.
- Low and high temperature results allowed us to contrast wettability alteration and interfacial dynamics effects.

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## ABSTRACT

In this study, low-salinity waterflooding enhanced-oil recovery mechanisms linked to the water–crude oil interfacial viscoelastic response are investigated considering a distinct interfacial effect, namely the water–crude oil interfacial viscoelasticity response. In contrast, wettability alteration is interpreted through analysis of capillary hysteresis. A capillary pressure experimental setup used in our previous research, is utilized here to capture capillary hysteresis. Moreover, new improvements over the traditional quasi-static porous plate method have been implemented to accelerate measurements. The dynamic-static method, i.e. a combination of continuous injection in drainage and stepwise quasi-static method in imbibition on short (< 1" long) core samples, was found to capture the correct envelopes of the capillary pressure curves in a much shorter time span. Two pairs of experiments were conducted to investigate the interfacial visco-elasticity and wettability alteration effects on capillary hysteresis. One pair is conducted on Minnelusa formation rock samples and diluted TC crude oil at 30 °C and without any significant aging to minimize wettability alteration. Two core plugs were flooded with high-salinity and low-salinity brines, separately. Results show that low-salinity brine can still increase oil recovery even in the absence of wettability alteration. This is attributed to the formation of a more visco-elastic brine–crude oil interface upon exposure to low-salinity brine, leading to a more continuous oil phase. Two additional experiment pairs were conducted on Berea and TC oil at high temperature (70 °C). Two cores were subject to the same experimental conditions except that one was allowed to age with crude oil for 3 weeks. This aging process was found to render the rock more oil wet, resulting in more pronounced capillary hysteresis and less oil recovery. Our study shows effects of two mechanisms, i.e. fluid–fluid interfacial visco-elasticity and wettability alteration, separately. The interpretation of our unique capillary pressure datasets sheds light on low-salinity waterflooding mechanisms.

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## 1. Introduction

Two of the most important features of multiphase flow in porous media are the presence of interfaces between immiscible fluid phases and the contact line where all of the interfaces intersect at the microscale. The fluid–fluid interface is commonly represented through a meniscus having a certain curvature that is compatible with capillary pressure jumps. Associated with this phenomenon,

the pore-scale capillary pressure is defined by the Young–Laplace equation as follows:

$$p_c = \sigma \left( \frac{1}{r_x} + \frac{1}{r_y} \right) \quad (1)$$

where  $p_c$  is the microscale capillary pressure,  $\sigma$  is the interfacial tension,  $r_x$  and  $r_y$  are the meniscus principal radii of curvature. Thus, capillary pressure depends on the pore dimension and the interfacial tension. The capillary pressure expressed in Eq. (1) is generally assumed to be valid regardless of whether the interface is moving or not. The pressures on the two opposing sides of the interface are

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different and under static conditions, the pressure difference equates the capillary pressure value:

$$p_c = p_n - p_w \quad (2)$$

where  $p_n$  is the non-wetting phase pressure and  $p_w$  is the wetting phase pressure. However, the relationship between capillary pressure and the pressure difference is not an intrinsic property of the system; it is dependent upon flow conditions. Hassanizadeh and Gray [1] derived the relationship between  $p_c$  and  $p_n - p_w$  from the force balance in the direction  $N$  normal to the interface as follows:

$$p_n - p_w = p_c + N \cdot (\tau_n - \tau_w) \cdot N - \nabla^\sigma \cdot \tau_{wn} \cdot N \quad (3)$$

where  $\tau_n$  and  $\tau_w$  are the viscous Newtonian stress tensors for non-wetting and wetting phases, respectively;  $\tau_{wn}$  is the viscous Newtonian stress tensor for the interface. Thus the relationship  $p_c = p_n - p_w$  is valid only under static conditions; for dynamic conditions,  $p_n - p_w$  will not be equal to the capillary pressure and will depend on the flow velocity. Sheng and Zhou [2] found the following relationship:

$$p_n - p_w = \sigma \left( \frac{1}{r_x} + \frac{1}{r_y} \right) \pm B \left( \frac{\mu q}{r} \right)^A \quad (4)$$

where  $\mu$  is the viscosity,  $q$  is the velocity,  $A$  and  $B$  are coefficients.

The aforementioned discussions are all at the microscale. At the macroscale, Bear and Bachmat [3], and Bear and Berruijt [4] defined the capillary pressure by analogy with the microscale capillary pressure, which is expressed in Eq. (5).

$$P_c = P_n - P_w \quad (5)$$

where  $P_c$  is the macroscopic capillary pressure,  $P_n$  is the average non-wetting phase pressure and  $P_w$  is the average wetting phase pressure. Consequently, the phase pressure is averaged over a portion of volume. Under dynamic conditions, the average phase pressure difference  $P_n - P_w$  depends on the time derivative of saturation. Hassanizadeh and Gray [5], and Kalaydjian [6] derived the equation based on thermodynamic theory:

$$P_n - P_w = P_c - \tau \frac{\partial S_w}{\partial t} \quad (6)$$

where  $\tau$  is a non-equilibrium coefficient, which is a nonlinear function of saturation and controls the rate at which equilibrium is reached. If the value of  $\tau$  is small,  $P_n - P_w$  and  $P_c$  will become instantaneously equivalent after the system equilibrium is disturbed. Thus,  $\tau$  acts like a capillary damping coefficient. If Eq. (5) is applied in a dynamic system, it is assumed that the disturbances can be eliminated instantaneously, which may only occur in high permeability porous media. Thus, Eq. (6) becomes necessary to correctly describe a dynamic system.

The underlying mechanisms for the dynamic effects are still unclear. Wildenschild et al. [7] have summarized the following mechanisms based on the observance of their water–air experiments:

- Water entrapment. At high flow rate during drainage, water in small pores can be isolated by draining its surrounding pores, thus causing higher water saturation under dynamic conditions. Moreover, the higher the flow rate, the less chances for all the pores to be drained.
- Pore water blockage. When a sudden and large pressure differential is applied to a fully saturated soil sample, the pores at the bottom are drained more quickly than the overall soil sample and are filled with air. This impedes further drainage and water stays behind.
- Air entrapment. When the water saturation is high during drainage, the air continuity could be poor if the sample holder is open to air only at the top. Thus, if air may be unavailable to

replace water, there will be higher saturation under dynamic conditions.

- Dynamic contact angle effect. It is known that in single capillaries, compared with static conditions, the dynamic contact angle is smaller in drainage and larger in imbibition [8]. Thus, this could cause higher capillary pressure in drainage and lower capillary pressure in imbibition.

In the context of our work, “static capillary pressure” (denoted as  $P_c^{stat}$ ) is used to refer to the intrinsic capillary pressure  $P_c$ ; “dynamic capillary pressure” (denoted as  $P_c^{dyn}$ ) will refer to the phase pressure difference  $P_n - P_w$  measured under dynamic conditions. Then, Eq. (6) becomes:

$$P_c^{dyn} - P_c^{stat} = \tau \frac{\partial S_w}{\partial t} \quad (7)$$

Many models have been proposed to relate macroscale capillary pressure to the wetting phase saturation,  $P_c = f(S_w)$ . These models implicitly lumped all the effects and processes that influence the equilibrium distribution of fluids, such as interfacial tension, rock wettability, pore size distribution and fluid–fluid interface. Moreover, it is commonly believed that when the wetting phase saturation reaches its residual value, the macroscopic capillary pressure goes to infinity. Since the capillary pressure is usually indirectly measured by the phase pressure difference in the external reservoirs bordering the porous media, it has been commented that the infinite capillary pressure at the residual wetting phase saturation is an inadequate statement [9,10], because the wetting phase at residual saturation is not continuous and loses hydraulic connection with the external reservoir.

One of the important features of the macroscopic capillary pressure–saturation relationship is its hysteretic character, which refers to the irreversibility or saturation path dependence. The source of hysteresis corresponds to (1) contact angle hysteresis, which refers to the advancing contact angle (in imbibition process) being larger than the receding contact angle (during the drainage process); (2) trapping of the non-wetting phase; (3) wettability alteration after a rock is contacted with crude oil, especially at a high reservoir temperature [11]. According to Johnson and Dettre [12], and Adamson [13], there could be three sources for contact angle hysteresis: surface roughness, heterogeneity and immobility at macromolecular scale.

Fluid trapping is caused by the fluid–fluid interface instability and rupture. In general, there are two types of interfaces (convex interface and selloidal interface). A convex interface refers to one with only positive elements of curvature and the movement of a convex interface in a pore or throat is related to piston-type displacement. A selloidal meniscus has components of positive and negative curvatures with radii on opposite sides of the meniscus. As the convex and selloidal interfaces move, they may exceed the limit of stability and will likely be ruptured. Then, the non-wetting phase could become disconnected by snap-off and break-off. Snap-off mainly occurs in the imbibition process, and it always involves the selloidal interface. As the wetting phase moves through a water film to a growing collar, at a critical capillary pressure, the interface will become unstable and rupture. Wettability and the topology are the determining factors for snap-off [14,15]. Break-off refers to the rupture of a convex interface by a jump through a pore and contact with an opposing pore wall in a piston-type displacement [16]. Buckley [17] suggests that rock wettability could be altered by crude oil through four different mechanisms: (1) Polar interactions. It predominates, if water is not present. Influencing variables include clay type, nitrogen, oxygen and sulfur content in oil. (2) Surface precipitation. It happens if the crude oil is a poor solvent with respect to its asphaltenes.

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