WETTABILITY EFFECTS ON OSMOSIS AS AN OIL MOBILIZATION MECHANISM DURING LOW SALINITY WATERFLOODING

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ABSTRACT

This experimental study identifies osmosis as an oil mobilization mechanism during low salinity waterflooding using capillary tubes and micromodels with different wetting properties. Oil-wet capillary tube tests verify that oil acts as a semi-permeable membrane for water transport when separating brines of different salinity. Osmotic oil mobilization was investigated using two-dimensional silicon-wafer micromodels for direct optical visualization of fluid interactions. Water diffusion was identified as the main transport mechanism at strongly water-wet conditions and capillary equilibrium. Osmosis was observed at both strongly water-wet and oil-wet conditions influenced by the pore-level distribution of oil and water. At strongly water-wet conditions osmosis displaced oil by expanding water-in-oil emulsions. At oil-wet conditions low salinity water was transported by osmosis to otherwise inaccessible regions of high-saline brine. Osmotic oil mobilization is a fluid-fluid interaction and should therefore be present in both sandstone and carbonate reservoirs. The experimental results support this and provide evidence of osmotic water transport and oil mobilization under various wettability conditions.

INTRODUCTION

Low salinity waterflooding (LSW) denotes injection of diluted brine concentrations in either secondary or tertiary recovery mode to increase oil recovery. Whereas conventional waterflooding uses formation brine or seawater predominantly to maintain reservoir pressure, LSW improves microscopic sweep by injecting water with diluted salt concentrations [1]. LSW has been reported to enhance oil production in a series of coreflood experiments [2], and has been tested in various field operations, see *e.g.* [3]. There is a consensus that an optimal wetting preference exists at which LSW is favorable for oil recovery. LSW is generally expected to shift reservoir wettability towards more water-wet conditions.

The initial research on LSW emphasized on fluid-rock interactions and the mechanisms that alter wettability of a rock surface. Coreflood results in sandstone provided a variety of theories aiming at defining the low salinity effect (LSE), as summarized by Sheng [4]. One of the more accepted hypotheses is the concept of multi-component ion exchange

(MIE), where inorganic Ca^{2+} and Mg^{2+} cationic exchange, between the negatively charged sandstone and the injected low salinity water, replace organic polar components at the rock surface to shift wettability towards water-wet [5]. The influence of cationic exchange was partially confirmed by others, who also reasoned that a major contribution was reduction of ionic brine strength [3]. Reduced ionic strength lowers the cationic screening potential that result in electrostatic repulsion and expansion of the electrical diffuse double layers surrounding clay and oil particles. Oil is desorbed from the surface and the initial wetting state is altered [3].

When exposed to LSW, and contrary to the established hypotheses [3, 5], an increase in recovery is also demonstrated in carbonates [6, 7]. Carbonate reservoirs are preferentially oil-wet due to a net positively charged surface [8]. According to Ligthelm *et al*. [3], reducing the ionic brine strength should yield greater attraction between organic polar components and the positive carbonate surface causing stronger oil-wet conditions, not a shift towards water-wet. The capability for multi-component cationic exchange is also restricted considering the positively charged rock. The general understanding is therefore that the underlying mechanism of LSE is yet to be found or that various mechanisms exist for different crude-oil/brine/rock systems [4, 9, 10].

Osmosis had been largely overlooked as a plausible mechanism for improved oil recovery during LSW [10]. Osmosis is a thermodynamic driving force for water transport caused by a difference in chemical potential between two aqueous solutions separated by a semi-permeable membrane [11]. In their initial work, Sandengen and Arntzen [10] observed oil displacement when separating high –and low-saline solutions by an oil membrane. A further investigation using oil-wet and water-wet micromodels showed that the osmotic effect was influenced by water connectivity and it was believed that osmotic gradients could not be established at water-wet conditions [12]. More recently, however, it was found that osmosis may be active as a transport mechanism in water-wet systems [13].

Current experimental data, obtained in micromodels, support the theory of osmosis as a mechanism for pore-scale oil mobilization regardless of the crude-oil/brine/rock system. Osmosis may therefore contribute to oil mobilization in both sandstone and carbonate reservoirs. The experimental analysis herein confirm this hypothesis and compose a collective theory identifying osmosis as one of the contributing mechanisms for oil mobilization at pore-scale during LSW.

EXPERIMENTAL

Previous work on osmosis using water-wet capillary tubes could not clearly identify if water transport occurred through the oil bulk or by film-flow induced by interfacial tension [10]. In this study, oil-wet PTFE tubes are used to isolate and verify water transport by osmosis through the oil bulk. In addition, silicon-wafer micromodels replicating the two-dimensional pore-structure of natural sandstone are used to qualitatively observe fluid-fluid interactions when performing wettability studies on osmotic oil mobilization during LSW. The strongly water-wet nature of the models is ideal for investigating osmosis by isolating known processes that occur during LSW *i.e.* wettability alteration towards more water-wet.

Fluids

Synthetic high-saline brines were made by adding given quantities of analytical-grade chemicals to deionized water (DIW), while pure DIW was used as low-saline brine. Oils were either toluene or crude oil (from a North Sea chalk reservoir). Toluene was used because of high water solubility to accelerate flux during osmosis. Colorants such as Oil Red O (red), Erinoglaucine disodium salt (blue) and Quinoline Yellow WS (yellow) were added at a concentration of ~0.05 wt% to optically distinguish the fluids at pore-scale by microscope. An overview of the fluids used and their characteristics are given in **Table 1**.

Table 1 – Chemical composition and characteristics of brines and oil phases.

Fluid ID	Composition	Ionic strength1) [M]	Osmotic pressure2) [bar]	Standard molar entropy3) [J/mol]	Interfacial tension w/Oil A [mN/m]
Brine A	5.0 M NaAc	5.0	124	138.2 (s)	14.0
Brine B	5.0 M NaCl	5.0	124	95.06 (aq)	25.3
Brine C	5.0 M $CaCl_2$	12.5	124	123.87 (aq)	22.5
Brine D	1.0 M NaCl	1.0	25	95.06 (aq)	20.4
DIW	Deionized water	0	0	69.95 (aq)	N/A
Oil A (Toluene)	C_7H_8	N/A	N/A	N/A	N/A
Oil B (Crude)	53 wt.% HC 35 wt.% aromatics 12 wt.% resins 0.9 wt.% asphaltenes	N/A	N/A	N/A	N/A
HMDS	>97% purity	N/A	N/A	N/A	N/A

1) Assume complete dissociation [14].

2) Assume non-electrolyte by Van't hoff equation [14]. Provides the maximum osmotic pressure that can develop in a solution when separated from deionized water.

3) The entropy contribution from one mole of substance at standard conditions [15].

Oil-wet capillaries for verifying osmotic water transport

To verify osmotic water transport a series of capillary tube tests were performed using oil-wet polytetrafluoroethylene (PTFE) tubes. The oil-wet nature restricts film flow along the surface, and water transport should occur only through the oil-phase by the osmotic gradient. Oil A was injected into the tube followed by high-saline Brine A and DIW on either side. A baseline without an osmotic gradient was also established using two water droplets of high-saline Brine A. The PTFE tubes were plugged and placed in an oven at 50 ºC for 20 days. Droplet movement was monitored and pictures were taken to document the process.

Micromodels for studying wettability effects on osmotic oil mobilization

Silicon-wafer micromodels representing natural sandstone were used to investigate the potential for osmotic oil mobilization by LSW at various wettability conditions. The strongly water-wet micromodels replicate a porous network in connection to adjacent channels acting as fractures. One micromodel was prepared using high-saline Brine D as connate water and Oil B as the oil-phase. At strongly water-wet conditions the high-saline brine adhered to the pore walls and was continuous throughout the porous network. Another system was prepared to represent oil-wet conditions. Here, Brine C was used as the high-saline phase and Oil A as the oil-phase. The micromodels were rendered oil-wet using Hexamethyl-diasilazane (HMDS) according to the modification procedure described elsewhere [12].

When preparing the micromodels they were 100% saturated with Brine C or D as connate water. Oil drainage was performed using either Oil A or B. After drainage, secondary waterflooding was conducted using high-saline brine allowing spontaneous imbibition to act as the main driving force until capillary equilibrium was reached. Baseline tests were run prior to performing LSW through the adjacent channel establishing a salinity contrast to the matrix high-saline brine. LSW was maintained at a flow rate of 1ml/day to represent dynamic conditions through a fracture network with water transport to the matrix dominated by small-scale diffusion processes.

A microscopic setup was used for pore-level visualization. Experiments were performed at ambient temperature and pressure with the micromodels placed horizontally on the microscope platform. Syringe pumps were connected to fluid distribution ports for brine and oil injection. Field-of-view was carefully selected showing parts of the matrix and adjacent flow channel. A camera and computer with associated imaging-software connected to the microscope captured time-lapse-pictures for documentation and post-image processing. LSW lasted between 7 to 14 days for each test.

RESULTS

Osmotic water transport in oil-wet capillary tubes

The effect of a salinity gradient on water transport through a semi-permeable oil membrane using oil-wet capillary tubes is shown in **Figure 1**. The two image sequences show water transport of low-saline water (yellow) through the oil-phase (red) into the adjacent high-saline solution (white) over time. With a salinity gradient present (image 1), all of the low-saline water was transported through the oil-phase into the high-saline brine after 20 days. The increased volume and color-shift indicates the direction of water transport from the low salinity water to the high-saline brine. Without a salinity gradient water transport was not observed (image 2), although some volume changes occurred due to slow dissolution of water in toluene causing the high-saline droplets to shrink.

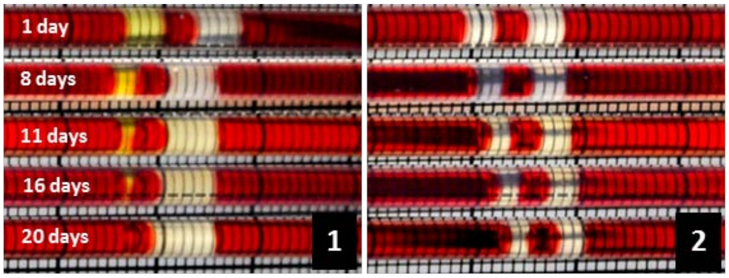

Figure 1. Osmotic water transport in oil-wet capillary tubes. The yellow phase is low salinity water, the red is the oil-phase and white is the high-saline brine. Time lapse images (1, 8, 11, 16 and 20 days) are shown from top to bottom. **1)** Osmotic gradients cause the low salinity water (yellow) to shrink as water is transported through the oil phase into the high-saline solution. **2)** The baseline result (*i.e.* high-saline solution only) showed no osmotic water transport only a slight dissolution of water into toluene**.**

The interfacial tension (IFT) between oil and water varies with salinity, and decreases upon dilution until reaching a minimum IFT at a critical salt concentration [16]. Hence, when two solutions with different salinity are separated by an oil-phase, an uneven balance is created across the oil-phase that may lead to convective water transport. IFT was measured for a combination of brines with Oil A (*cf.* Table 1). To determine water transport as an effect of osmosis only, combinations of Brine A, B and DIW were used in capillary-tube tests. Brine A has a lower IFT compared to Brine B and behaves qualitatively different in terms of capillary forces, but the two brines have the same ionic strength and osmotic gradient (*cf.* Table 1).

Table 2 provides an overview of the capillary tube tests performed with configurations of high – and low saline solutions separated by oil. Expansion of the reference solution is defined by "+", contraction by "-" and no change is termed "0". In all cases the observed results matched the direction of water transport predicted theoretically by osmosis.

Table 2 – Water transport in oil-wet capillary tube tests. The experimental results are correlated with theoretical predictions from interfacial tension and osmosis.

Reference solution	**Opposing solution**	**Theoretical Prediction Osmosis**	**Theoretical Prediction Interfacial tension**	**Experimental Observation**
Brine B	Brine B	0	0	0
Brine B	Brine A	0	-	0
Brine B	DIW	+	-	+
DIW	DIW	0	0	0
Brine A	DIW	+	+	+
Brine A	Brine A	0	0	0

Wettability effects on osmotic oil mobilization

Oil mobilization during LSW occurs in response to forward osmosis when a system is partially saturated with high-saline brine and oil [17]. Both strongly water-wet and oil-wet silicon-wafer micromodels were used to evaluate the influence of wettability on osmotic oil mobilization. To create a salinity contrast to the high-saline connate brine

LSW was injected through the adjacent fracture only. No fluid displacement was observed in baseline experiments, where the salinities of injected and connate brine were equal.

Strongly water-wet conditions

At strongly water-wet conditions the initial high-saline brine adhered to the pore walls creating a continuous water layer. Due to water connectivity and ceased capillary imbibition the main transport mechanism was water diffusion from the low salinity phase through film-flow diluting the high-saline connate brine. The increasing water-volumes within the porous network resulted in a redistribution of fluids and capillary trapped oil was viscously displaced into the fracture to give way for the preferred water phase. **Figure 2** shows how diffusion of low salinity water mobilized oil in a strongly water-wet system.

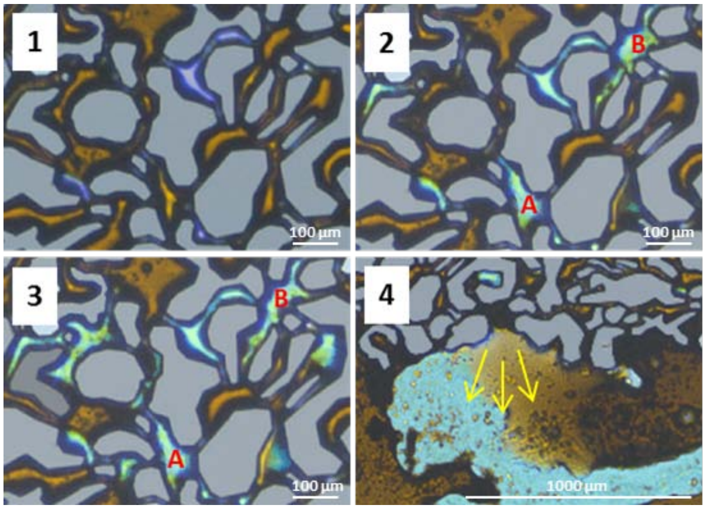

Figure 2. Low salinity water diffusion and oil mobilization. Oil B (crude oil, brown), Brine D (blue), DIW (turquoise) and rock grains (grey). Image 1-3 shows several pores, and Image 4 shows the fracture located below (different length scale). **1)** Initial fluid distribution. **2)** Diffusion of DIW through pore A and film-flow into pore B displacing oil to adjacent pores. **3)** Continued DIW oil displacement from several interconnected pores. **4)** Fluid redistribution leads to oil displacement into adjacent fracture (yellow arrows).

The osmotic effect was observed in small high-saline brine-droplets dispersed within the oil-phase indicated by the yellow arrows in **Figure 3**, image 1. These water-in-oil emulsions started to grow due to osmotic water transport during LSW and accumulated into larger emulsions by coalescence and Ostwald ripening. This growth was not observed without a salinity gradient (*i.e.* baseline). The continuous aggregation (image 2) embodied most of the oil-phase, slowly displacing it towards the lower channel and into connecting pore throats.

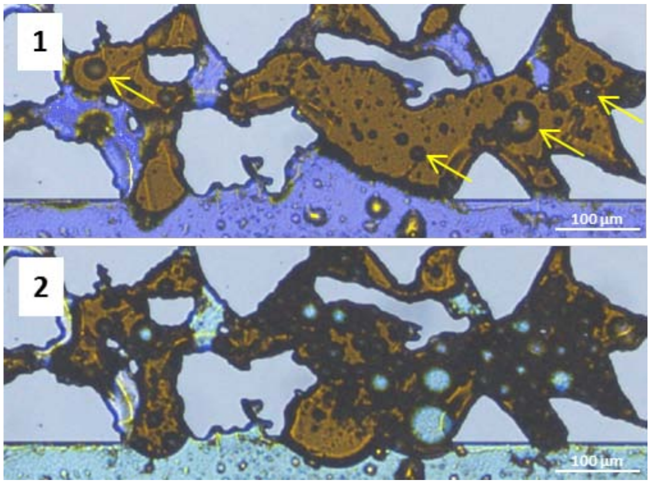

Figure 3. Oil displacement by osmotic water-in-oil expansion. Oil B (crude oil, brown), Brine D (blue), DIW (turquoise) and rock grains (grey). **1)** The fluid distribution prior to performing LSW. An oil-phase is located in the pore network in contact to the adjacent channel. Dissolved droplets of high-saline brine within the oil are indicated by the yellow arrows. **2)** During LSW, the water-in-oil emulsions start to grow and the aggregation of emulsions displace the oil-phase.

Oil-wet conditions

At oil-wet conditions, osmotic gradients were established when the low salinity water contacted the oil-phase leading to swelling of capillary trapped high-saline brine. **Figure 4** shows expansion of capillary-trapped brine (blue) and subsequent oil (orange) mobilization by osmosis. As the capillary trapped connate brine expanded the oil was displaced towards the flow channel as indicated by the yellow arrows in image 2. The oil-phase was mobilized into the fracture when the viscous pressure from brine expansion exceeded the capillary pressure withholding oil in matrix (image 3).

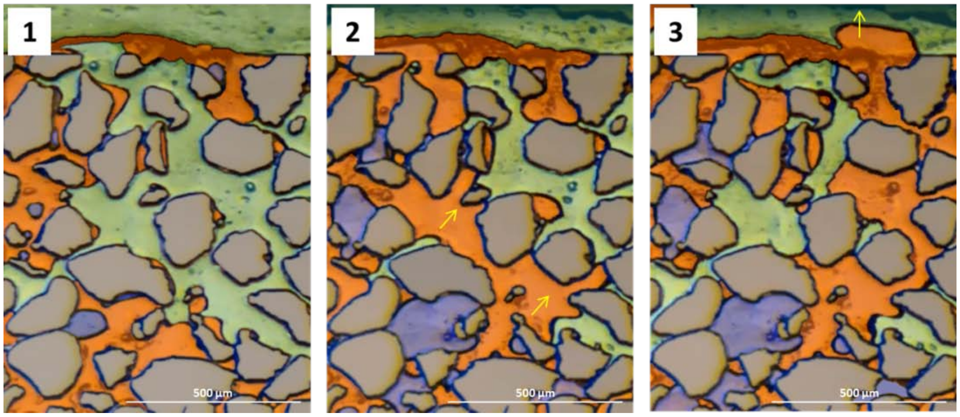

Figure 4. Osmotic oil mobilization by capillary- trapped brine expansion. Oil A (orange), Brine C (blue), DIW (turquoise) and rock grains (grey). **1)** Fluid configuration after initiating LSW. Capillary trapped regions of high-saline brine are found within the oil-phase. **2)** The high-saline regions start to expand in response to osmotic induced water transport mobilizing oil towards the fracture by the yellow arrows. **3)** Oil is produced from the porous medium and into the nearby flow channel.

DISCUSSION

Verification of osmotic water transport in capillaries

The oil-wet capillary tube tests represent a situation where stagnant pockets of connate water, either through snap-off or bypassed, are surrounded by oil. The direction of water transport observed in **Figure 1** matched prediction by osmosis, and suggests limited influence of salinity effects on IFT. This is also corroborated by previous qualitative observations in water-wet micromodels [10]. Rate of oil mobilization by osmosis depends, among other factors, on the thickness of the oil-phase. It can be compared to the diffusion rate by Fick's law, given the diffusion coefficient by the Stokes-Einstein (SE) equation. SE assumes that water diffusion is inversely proportional to the oil-phase viscosity. However, when water moves through an oil-phase without hydrogen bonding the smaller water molecules diffuse faster than predicted [18], and additional considerations are required to describe the waterflux.

The pore-scale osmotic effect observed in capillary tubes is relevant during spontaneous imbibition, where oil production is determined by capillary forces without viscous flow. It is believed that the osmotic effect may contribute favorably during core-scale spontaneous imbibition of low salinity water by expanding volumes of high-saline brine and mobilizing oil out of the core.

Verification of water diffusion in micromodels

Water diffusion was observed as the main transport mechanism at strongly water-wet conditions by the connectivity between fracture bulk-flow and matrix brine. The transport is induced by the chemical potential gradient between the high- and low-saline phases. The potential is defined as the sum of Gibbs free energy for all components in the solution. When salts are mixed with water to form high-saline brine the components spontaneously dissolve into electrically charged ions due to the resulting increase in entropy [11] (*cf.* Table 1). Heat is released during the dissolution process and the Gibbs free energy of the solution, *e.g.* the energy available to do work, is reduced. Low salinity water (DIW in this work) has a higher chemical potential than high-saline brine, and water will move from regions of high to low potential as observed in **Figure 2**. The system is stabilized when Gibbs free energy reaches a minimum for the system as a whole. Hence, water will diffuse to re-establish thermodynamic stability.

Oil mobilization by water diffusion

It was mainly the diffusion of solvents (*i.e.* water) that diluted the high-saline brine in matrix to equilibrate the difference in chemical potential. Ion-dipole forces between salt ions and water molecules reduce the diffusion of solutes as they are stronger than the induced hydrogen bonds between water molecules [19]. Water diffusion was initially identified as a dilution mechanism causing film-expansion and droplet growth during LSW [13]. It was reasoned that the accumulation of water in local water-films increased the water pressure leading to viscous oil displacement into nearby flow channels. Research by Stoll *et al.* [20] emphasized that in absence of significant viscous and capillary forces, wettability modifiers must initially diffuse under its own concentration gradient into the matrix to activate its chemical process. The results in **Figure 2** support these findings and illustrate how propagation of water may re-distribute the fluids in matrix, displacing oil from pore-to-pore and towards the fracture.

Verification of osmotic water transport in micromodels

Oil mobilization was observed at both strongly water-wet and oil-wet conditions, with water transport to otherwise inaccessible regions of high-saline brine and water-in-oil emulsions (**Figure 3** and **4**). Osmosis is the transport of water when high- and low-saline brines are separated by a semi-permeable membrane that is permeable to pure water, but impermeable to ions [11]. To balance the chemical potential gradient, *e.g.* osmotic gradient, the solvent must have equal values in the two brines by having the two brines at different pressures [14]. Hence, water is transported from the low-saline brine through the membrane to compensate for the reduction in chemical potential of the high-saline brine. The osmotic pressure within the high-saline brine increase with addition of water molecules, and transport stops when the osmotic pressure balances the flow induced by the osmotic gradient.

Semi-permeable membrane characteristics of oil

The semi-permeable properties of oil depend on the intermolecular forces acting between the solute, solvent and hydrocarbon compounds present. The capacity for oils to bind with hydrogen depends on the hydrocarbon composition. Mineral oils with chains of alkanes have zero dipole moment and hence no capacity to hydrogen bond [18]. Asphaltenes and some aromatics present in most crude oils have structures to create weak hydrogen couplings that influence the flux of water [21, 22]. Hence, an oil-phase with a higher fraction of non-polar alkanes should have better membrane properties and act as a more efficient conduit for water molecules under the influence of osmotic gradients.

Wettability effects on osmotic oil mobilization

At strongly water-wet conditions, isolated high-saline brine existed only as water-in-oil emulsions that expand during LSW by aggregating inside the phase. Researchers have drawn special attention to the growth of water-in-oil emulsions: some explain it as a result of surface-active components arranging micelles transporting water molecules to inaccessible brine, see *e.g.* [23], whereas others have suggested that the water transport is caused by osmosis [15-13]. In addition, it has been shown that water-in-oil emulsions can grow without the presence of surface-active components by using mineral oil during LSW [15]. It is therefore suggested that the growth of water-in-oil emulsions is a result of osmosis, supported by results from the present (**Figure 3** and **4**) and previous work [13].

Water-in-oil emulsions are present in this work independent on wettability. The stability of water-in-oil emulsions depend on the composition of the oil-phase. Oil B, *i.e.* crude oil, contains asphaltenes that help stabilize emulsions [24]. Mineral oils containing non-polar alkanes, as used in the oil-wet system, are not interfacially active and cannot form stabilized films around the water-droplets at the oil/water interface [24]. As long as there are compounds present that can form stable films, emulsions should develop.

An oil-wet system represents the most ideal for osmosis as it holds a number of suitable membranes and confined volumes of high-saline brine for oil mobilization (**Figure 4**). Several isolated regions of high-saline brine started to expand and the fluids within matrix were redistributed as observed in images 1-2. As a result oil was mobilized into the nearby flow-channel (**Figure 4**, image 3).

The calculations of osmotic pressure in Table 1 indicate a high potential for viscous pressure build-up during LSW. These theoretical values are unlikely to develop within the pore-space due to the imperfect membrane properties of the oil. In corefloods the reported pressure response is usually transient or modest at best when injecting low salinity water [1, 7, 10, 25]. If osmosis is a significant contributor to the overall trend one would observe a general increase in pressure followed by a slow decrease as the gradient is gradually balanced, see *e.g.* [25]. However, a relationship cannot be drawn between the pressure gradients reported in literature and osmosis.

Osmosis as low salinity effect on reservoir scale

LSE is more relevant at oil-wet conditions as the general understanding is that LSW shifts wettability towards more water-wet increasing oil recovery. However, it has been documented that LSE improves oil recovery without the necessary conditions, see *e.g.* [1, 25]. The variety of circumstances under which LSE may or may not be observed suggests that more than one mechanism may be in play [1]. In the present work, osmosis is observed independent on oil/brine/rock interactions and should therefore be applicable to both oil and water-wet systems.

In a highly flooded water or mixed-wet reservoir it is suggested that enclosed volumes and emulsions of connate water will swell in response to osmosis. Re-mobilization of oil will occur at pore-level until the oil-films either ruptures or thermodynamic stability is re-established. However, it is not likely that the effect will greatly reflect on total recovery. A fractured, oil-wet reservoir (*i.e.* **Figure 4**) would be an ideal system for osmosis to have a significant impact on oil recovery. In a fractured system osmosis will act as a driving force for water transport into the oil-wet matrix. Osmosis may contribute positively to open pathways that increase the water relative permeability, aiding other effective low salinity mechanisms to take place, such as wettability alteration in otherwise inaccessible regions of the matrix, and for oil to be produced into high permeable flood zones.

As osmosis occur independent on wettability is likely to be present in both sandstone (water-wet) and carbonate (oil-wet) formations. To properly determine what effect osmosis will have on recovery, one could use model systems where wettability alteration is excluded. This could help to better quantify the extent of osmosis during LSW. The present work identifies osmosis as a pore-level LSE that improves microscopic sweep in combination with other low salinity mechanisms.

CONCLUSIONS

- Oil-wet capillary tube tests verified osmotic water transport as oil-bulk diffusion from a solution of low salinity to a solution of high salinity due to the semi-permeable characteristics of oil.
- The work performed substantiate osmosis as an underlying low salinity effect for oil mobilization at pore-level by identifying osmotic water transport at both strongly water-wet and oil-wet conditions during low salinity waterflooding.
- Osmotic water transport mobilized oil by either swelling water-in-oil emulsions or expanding otherwise inaccessible regions of capillary trapped high-saline brine.
- At strongly water-wet conditions and capillary equilibrium water diffusion by film-flow was the main transport mechanism for oil mobilization.
- The osmotic effect is influenced by fluid distribution of oil and water. It is independent of system wettability and should therefore be present as a pore-scale oil mobilization mechanism in both sandstones (*e.g.* water-wet) and carbonates (*e.g.* oil-wet) during LSW.

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