SCA2008-42 1/12

WETTABILITY ALTERATION BY SPONTANEOUS IMBIBITION OF SULPHATE-CONTAINING WATER INTO CHALK CORES WITH DIFFERENT BOUNDARY CONDITIONS

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ABSTRACT

Seawater injection may improve oil recovery from the Ekofisk chalk reservoir and a lot of experimental studies have been reported for the spontaneous imbibition (SI) of sulphate-containing water into cylindrical chalk core plugs with all surfaces open to the imbibing fluid.

In this paper, the influence of the boundary conditions on SI of sulphate-containing water was investigated with formation water without sulphate as reference. Three different boundary conditions were studied: open top end, open bottom end and open both top and bottom ends. In all cases the cylindrical surface was closed. Also, a one-dimensional numerical model was used to simulate the tests.

For core plugs with both top and bottom ends open, SI tests were carried out at 50°C, the oil recovery from the top and the bottom was recorded separately. Higher oil recovery was observed from the top end since both countercurrent and cocurrent flow take place at the top, while only countercurrent flow at the bottom end. Gravity was observed as an important factor for cocurrent flow, demonstrated by the larger oil recovery gap between the top end and the bottom end for the longer core plugs.

The SI tests also showed a higher oil recovery for sulphate-containing water as the imbibing fluid than for formation water. At 130°C, with both top and bottom ends open, higher oil recovery was observed than for only one end open since a larger area of the core plug was exposed to the imbibing fluid. With only one end open, the imbibition rate was slower because the slow process of imbibing fluid entering further into the core plug from top or bottom. If only one end is open, top end or bottom end, it is difficult to get a consistent conclusion from oil recovery performance.

With sulphate as the wettability alteration agent, diffusion into the core plug influences the imbibition process and change the capillary pressure and relative permeabilities dynamically. This effect was investigated by a one-dimensional numerical simulator developed to analyze the influence of the boundary conditions. An experimental example

SCA2008-42 2/12

can be reasonably matched, and the simulation result showed that oil recovery for a core plug with only the top end open is higher than with only bottom end open.

INTRODUCTION

During SI in fractured reservoirs, both cocurrent and countercurrent flow can take place (Al-Lawati and Saleh, 1996; Hamon and Vidal, 1986), dependent on the ratio of the gravity to capillary forces, the water injection rates and boundary condition of the matrix block in the fracture network. Water may enter the block and displace the oil out cocurrently, or if the two phases flow in opposite direction, it is countercurrent flow. In the reservoir, conditions leading to cocurrent imbibition may be gravity segregation of oil and water in the fractures, whereby only parts of the matrix block surfaces is exposed to water. Countercurrent imbibition is the only displacement mechanism of a matrix block completely surrounded by water in the fractures.

Bourbiaux and Kalaydjian (1990) performed cocurrent and countercurrent spontaneous water-oil imbibition tests on single, vertical block samples of strongly water-wet sandstones at laboratory conditions. In the dominant cocurrent flow case, the measured brine saturation profile showed a brine-oil front moving regularly from the lower to the to the upper end, and the slope of this front decreased with time because the driving effect of gravity forces decreased as the front nearing the upper face. In the pure countercurrent case, the oil/brine front became flat as it progressed towards the bottom of the sample. In the combined co- and countercurrent case, two fronts progress at similar speed from each sample end towards the middle. The movement of both oil and water in opposite directions during countercurrent imbibition is a slower process than the cocurrent displacement.

Babadagli et al. (1999) performed both cocurrent and countercurrent imbibition tests with Berea sandstone samples. For the countercurrent experiments, as less matrix contact area is exposed to imbibing fluid, more time is required for the capillary imbibition process to start. All the cocurrent experiments lead to nearly the same ultimate oil recovery for all boundary conditions, but the recovery rate decreases as more surface area of the matrix is isolated. The effects of boundary conditions on the recovery rate are prominent.

A lot of studies were done for the sandstone cores with relatively high permeability of more than 100mD (Jadhunandan and Morrow, 1991; Reis and Cil, 1993), while chalk and diatomite have higher porosity and lower permeability. Cuiec (1994) did SI experimental work with low permeability chalk and stated that countercurrent flow is the dominant mechanism, particularly when the brine flow displaces oil from the bottom of the blocks. Zhou et al. (2001; 2002) did the countercurrent SI tests with diatomite. They found that countercurrent SI was more sensitive to core-scale heterogeneities compared with cocurrent flow. Standnes (2004) concluded that both cocurrent and countercurrent tests should be performed to evaluate the reservoir potential to predict the oil recovery rate and ultimate oil recovery more accurately.

SCA2008-42 3/12

The laboratory SI tests on cylindrical outcrop chalk cores with all surfaces open indicate that seawater helps to improve oil recovery from moderately water-wet chalk fields (Austad et al., 2005). It was observed that high temperature and the presence of sulphate ions in the injected seawater were the key factors for wettability modifications towards more water-wet conditions.

The purpose of this paper is to check how the boundary condition may influence the spontaneous seawater imbibiton into chalk cores, and to simulate and interpret the flow processes. Chalk cores in different wetting states caused by different oil types, different initial water saturations and different temperatures are prepared, and the effects of boundary condition on spontaneous imbibition are investigated.

EXPERIMENTAL

Materials

<u>Porous media</u> The chalk core plugs are from Stevns Klint (SK) nearby Copenhagen, Denmark. It is a soft and highly porous chalk of Maastrichtian age. The porosity is in the range of 45-50%, and permeability is about 2-5mD (Milter and Austad, 1996). It has a specific surface area of about 2m²/g (Røgen and Fabricius, 2002). It is assumed similar to the reservoir chalk in the Ekofisk field in the North Sea (Frykman, 1994).

 $\underline{\mathrm{Oil}}$ Two kinds of oils were used to prepare the core plugs for SI tests. They were made from one crude oil from the North Sea. The Oil A was prepared by diluting the crude oil with n-heptane in the ratio of 40/60 n-heptane/crude oil by volume, and then filtrated through a 5µm Millipore filter. The acid number (AN) was analyzed to be 2.07mgKOH/g. In preparation of Oil B, the Oil A was added 10 weight % silica and the mixture was stirred for 3 days. Another 10 weight % silica was added again and the mixture was stirred for 2 days to get oil B with AN about 0.17mgKOH/g. The oil was then centrifuged and filtered through a 5µm Millipore filter. Oil A and B was then mixed in the weight ratio A/B=1:5 to prepare Oil C with AN=0.49mgKOH/g. Oil A and Oil C were used during the core preparation.

<u>Brines</u> Synthetic Ekofisk formation brine (EF-brine) was prepared and used as the initial water present inside the core plugs. The synthetic seawater SSW were used as imbibing fluids, also EF-brine was used as the imbibing fluid which was a reference case. The brine components are shown in Table 1.

Procedures

Chalk cores plugs were drilled from the same chalk block and shaved to the expected dimensions. Before they were dried to constant weight, the possible initial sulphate inside the cores which could influence the wetting state was cleaned out. The cores were flooded at 50°C with 4PV distilled water from each direction in the Hassler core holder.. Cores LY1-2 and LS1-1 was used for the early experiment and did not experience this cleaning procedure. After all the core plugs were dried to constant weight they were prepared according to the method described by Standnes and Austad (2000):

SCA2008-42 4/12

• The cores were saturated with EF-brine after having been dried at 90°C to constant weight and evacuated. They were then flooded with 2PV of oil A or C in each direction to get water saturations in the range of 24.9% to 32.7 %.

- Lower water saturations around 10% was achieved by porous plate desaturation using strongly non-wetting nitrogen before flooding with oil.
- The core plugs were then aged in the same kind of oil as the oil used for flooding procedure at 90°C for about 30 days or at 50°C for about 5 days. A large amount of surface-active components from the oil may adsorb onto the outermost surfaces of the cores. Hence, after aging, the outermost 2 mm of each core was shaved off prior to the SI test to ensure a more uniform wettability of the core. SI tests were performed after sealing the core plugs with epoxy to get different boundary conditions, see Figure 1.

SIMULATION

In the previous work (Yu et al., 2008a; 2008b), a one dimensional model was built to study the wettability alteration caused by seawater spontaneous imbibition into preferentially oilwet chalk cores. The relative permeability and capillary pressure curves are dynamically changed according to the wetting state change, which is again controlled by the concentration of the wettability alteration agent having entered the core plug. For 1D countercurrent flow, conservation of mass, assuming incompressible flow, combined with Darcy's law, in dimensionless form, give

$$S_t + \gamma f_T(s, c)_x = \varepsilon \left[\left(B_1(s, c) S_x \right)_x + \left(B_2(s, c) C_x \right)_x \right], \tag{1-1}$$

$$\left[sc + a(c)\right]_{t} + \gamma \left[cf_{T}(s,c)\right]_{x} = \left(D(s)c_{x}\right)_{x} + \varepsilon \left[\left(cB_{1}(s,c)s_{x}\right)_{x} + \left(cB_{2}(s,c)c_{x}\right)_{x}\right], \tag{1-2}$$

where
$$\gamma = \frac{L\Delta\rho Kg}{\mu_r D_r}$$
, $\varepsilon = \frac{KP_{cr}}{\mu_r D_r}$, and $f_T(s,c) = [f\lambda_o](s,c)$,

 $B_1(s,c) = -K\lambda_o(s,c)f(s,c)(P_c)_s$, $B_2(s,c) = -K\lambda_o(s,c)f(s,c)(P_c)_c$, $\Delta\rho$ is the density difference between water phase and oil phase, K absolute permeability, g acceleration due to gravity, c concentration of wettability alteration agent, f fractional flow function, λ_o is oil mobility, P_{cr} , μ_r , D_r are reference capillary pressure, viscosity, and molecular diffusion coefficient, D(s) molecular diffusion coefficient, a(c) adsorption isotherm.

RESULTS AND DISCUSSION

SI tests at different boundary conditions

Eleven core plugs were prepared for the SI tests to evaluate the influence of boundary conditions. Core data are shown in Table 2. Two core plugs LY1-2, LS1-1 were aged at 50°C for 5 days, and the SI tests were performed at 50°C with boundary condition of two ends open, shown by (b) in Figure 1. The produced oil from top and from bottom were collected separately and oil recovery was recorded in Figure 2 and Figure 3. Oil recovery from top end was obviously higher than that from bottom end. From the bottom end, countercurrent flow took place due to capillary imbibition, against the force of gravity. For the top end, oil was recovered caused by capillary imbibition working with gravity.

SCA2008-42 5/12

Moreover, oil near the bottom end could be produced from the top, caused by gravity alone, that is, cocurrent flow occurred from the top end. The importance of gravity influence was confirmed by comparing Figure 2 to Figure 3, the recovery difference between top and bottom ends was decreased from 15% to about 7% with the core length decreasing from 7cm to 4cm.

Three core plugs LK11, LK12 and LK21 were prepared with the same oil as the core plugs LY1-2 and LS1-1 but with higher aging temperature, longer aging time and with cleaning procedure. Compared with Figure 2 and Figure 3, the lower oil recovery at 70°C showed in Figure 4 displays that the core plugs now are more oil-wet because high temperature and long aging time both contribute to oil-wetness, also lower initial sulphate content after cleaning procedure can help to get more oil-wet state (Puntervold et al., 2007). Shown in Figure 4, SSW can further improve the oil recovery over that of EF-brine. Taking SSW as the imbibing fluid, the oil recovery for core plug LK11 with only top end open was a little bit smaller than that for the core plug LK12 with only bottom end open, and it is difficult to conclude about the boundary effect in this case.

The SI tests at 130°C were performed with core plugs prepared with different boundary conditions, different initial water saturations and different initial water types. The core plugs were put into the steel cells surrounded by imbibing fluids, and 10 bars pressure was supported to avoid boiling. The results are shown in Figure 5 and Figure 6. Core plugs with initial water saturation S_{wi} of about 25% were treated to get different boundary conditions, (1) top end open, (2) both top and bottom ends open, and (3) bottom end open. For the case with both top and bottom ends open, the total oil production was recorded since the separate oil collection from top end and bottom end can not be realized with the present equipment. In Figure 5, with two ends open, more area was exposed to SSW, the oil recovery was higher than the case with only top end open. The oil recovery from 5 to 25 days kept increasing, probably due to the wettability alteration process caused by seawater entering the core plugs, as suggested by Yu et al. (2008b). For the cases with only top or bottom end open, shown in Figure 5 and Figure 6, the oil recovery from the bottom was slightly higher than that from the top, which needs the further investigation by preparing the samples with similar properties such as permeability, initial water saturation, etc.

The effect of initial water saturation was shown in SI tests at 130°C. Comparing Figure 5 and Figure 6, lower oil recovery was reached for the core plug with lower initial water saturation, with recovery 11% for Swi=9.1% and 16% for Swi=25.6%. This is consistent with the previous results (Puntervold, 2007) that showed that initial water saturation in chalk, which undergoes a wettability alteration process, had minor influence on the SI process. Secondly, the type of initial water affects the SI performance. We changed the initial water types present inside the core plug from EF-brine to SSW. In Figure 6, higher oil recovery was achieved by using SSW as the initial water to saturate the core plugs. For top end open, recovery changes from about 10% to 25%, and for the case with bottom end open, recovery changes from around 12% to 27%. It is very probably that the SSW decreases the contact of oil with the rock surface, which then leads to a more water-wet

SCA2008-42 6/12

condition compared to EF-brine as initial water. The work by Puntervold et al. (2007) showed that sulphate in the SSW may be the key factor to explain this phenomenon.

Simulations of 1-D countercurrent flow

The countercurrent flow for the core plugs with only the top end or the bottom end open was simulated with the 1D model. We assume the core plugs are in the same wetting state to start SI tests. Taking core plug LY2-5 as an example, the core properties were used as input data. The simulation results for two cases, top and bottom open, are shown in Figure 7.

For the case with top end open, the oil recovery curve in Figure 7 (a) can be compared to the experimental result in Figure 5, which was qualitatively matched. If the bottom end is open, the simulation results show a lower oil recovery than the case with top end open. This can be explained from the water saturation profile after 40 days SI in Figure 7 (b), where, along the x-axis, 0 denotes the sealed end and 1 the open end. For the case with the top end open, the water saturation (S_w) near the sealed end (core bottom) ~ 0.4 was higher than that near the open end (core top) ~ 0.31 . For the case with bottom end open, S_w at the sealed end (core top) ~ 0.27 was lower than that near the open end (core bottom) ~ 0.31 . Since water is the denser phase, it tends to accumulate near the core bottom due to gravity effects. This can explain the simulated oil recovery difference between top end open and bottom end open for each case. Compare S_w near the bottom end for these two case, the difference between ~ 0.4 and ~ 0.27 is due to the positive contribution of gravity on the countercurrent flow for the case with only top end open, while negative contribution for the case with only bottom end open, i.e., oil production from the bottom needs to overcome the buoyancy force. The gravity effect finally leads to the difference of oil recovery in Figure 7 (a), the balance between gravity and capillary force determines how much the difference is, and it also determines how much the S_w difference is distribution along the core plug.

CONCLUSION

By the 1D SI tests on cylindrical chalk cores, sulphate-containing water show potential for improving oil recovery. The initial water saturation, the oil types to age the cores and temperature of imbibing fluid are found important to control the wetting state. With the specific investigation on the effects of the boundary condition show that boundary condition is important for the SI process and should be considered to evaluate the oil recovery performance. With more area exposed to the sulphate-containing imbibing fluid, the oil recovery increased. For the case with only top end open, only countercurrent flow takes place, while for the case with top and bottom ends open, countercurrent flow takes place for both ends and at the same time cocurrent flow can exist for the top end. Together with the gravity influence, the oil recovery from top end is higher that that from bottom end. For the case with only one end open, top end or bottom end, which means the area exposed to the imbibing fluid is the same, a consistent trend cannot be concluded with the present results because the uncertainty of other influencing parameters.

SCA2008-42 7/12

By simulation, the different oil recovery processes were analyzed for the cases with only top end open or only bottom end open, the oil recovery for the case with bottom end open is lower. The combination of gravity and capillary forces determines the behavior.

ABBREVIATION

EF-brine Ekofisk formation brine
OOIP Original oil in place
SI Spontaneous imbibition
SSW Synthetic seawater

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SCA2008-42 8/12

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Table 1. Components of the brines.

Ion	EF-brine (mol/l)	SSW (mol/l)		
Na ⁺	0.684	0.450		
K ⁺	-	0.010		
Mg ²⁺	0.025	0.045		
Ca ²⁺	0.231	0.013		
Cl ⁻	1.196	0.525		
HCO ₃	1	0.002		
SO_4^{2-}	1	0.024		
TDS, g/l	68.01	33.39		

SCA2008-42 9/12

Table 2. Properties of core plugs used for the SI tests.

	Aging Aging	Core properties			SI						
		Temp.	time (days)	Diameter (cm)	Length (cm)	Porosity (%)	Swi (%)	Initial Water	Boundary condition	Temp. (°C)	Imbibing fluid
LY1-2	C	50	5	3.80	7.04	50.1	25.7	EF-brine	Two Ends Open	50	SSW
LS1-1	C	50	5	3.80	4.05	50.2	24.9	EF-brine	Two Ends Open	50	SSW
LK11	C	90	30	3.75	6.30	48.8	30.1	EF-brine	Top end Open	70	SSW
LK12	C	90	30	3.75	6.62	48.0	27.1	EF-brine	Bottom end Open	70	SSW
LK21	C	90	30	3.76	6.81	46.1	32.7	EF-brine	Top end Open	70	EF-brine
LT1-2	Α	90	30	3.70	4.98	50.3	25.6	EF-brine	Two ends Open	130	SSW
LY2-5	Α	90	30	3.70	4.54	50.7	25.6	EF-brine	Top end Open	130	SSW
#11	Α	90	29	3.75	6.90	48.4	9.1	EF-brine	Top end Open	130	SSW
#12	Α	90	29	3.75	6.50	47.2	8.5	EF-brine	Bottom end	130	SSW
#21	Α	90	29	3.74	6.99	46.9	8.0	SSW	Top end Open	130	SSW
#22	A	90	29	3.77	6.99	48.5	7.9	SSW	Bottom end	130	SSW

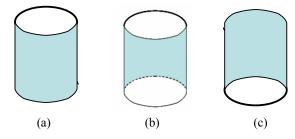


Figure 1. Schematic diagram of boundary conditions for the cores. (a) only top end open, (b) both top and bottom ends open. (c) only bottom end open.

SCA2008-42 10/12

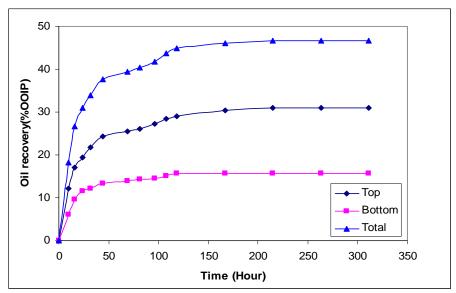


Figure 2. SSW SI into core plug LY1-2 with two ends open, S_{wi} =25.7%. Oil recovery from top and bottom ends was recorded separately. Tests were performed at 50°C.

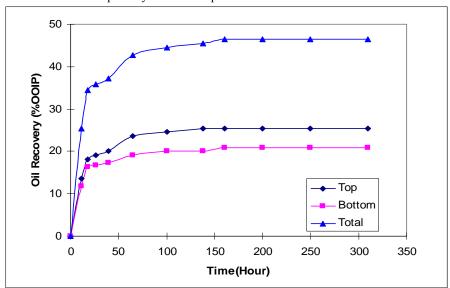


Figure 3. SSW SI into core plug LS1-1 with two ends open, S_{wi} =24.9%. Oil recovery from top and bottom ends was recorded separately. Tests were performed at 50°C.

SCA2008-42 11/12

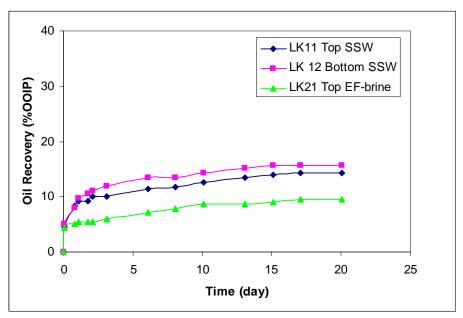


Figure 4. SI of SSW and EF-brine into core plugs with different boundary conditions, LY11 and LK 12 only top end open, LK21 bottom ends open, Swi 27-32%. Tests were performed at 70°C.

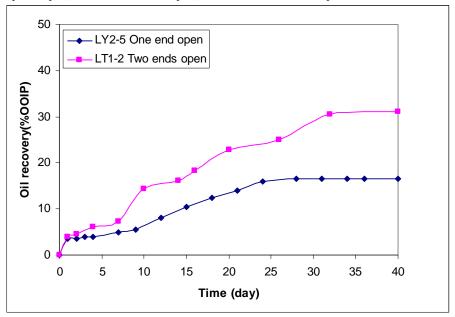


Figure 5. SI of SSW into core plugs with different boundary conditions, LY2-5 only top end open, LT1-2 both top and bottom ends open, S_{wi} about 25%. Tests were performed at 130°C.

SCA2008-42 12/12

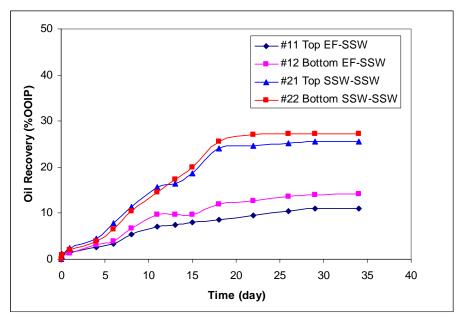


Figure 6. SI of SSW into cores with different initial waters and different boundary conditions, #11 and #12 have EF-brine as initial water, #21 and #22 have SSW as initial water. #11 and #21 only top end open, #12 and #22 only bottom end open, S_{wi} about 10%. Tests performed at 130°C.

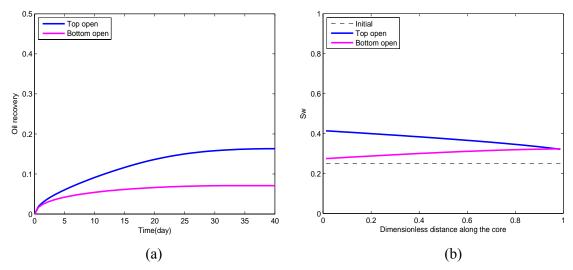


Figure 7. Simulation of SI test on core plug LY2-5 with different boundary conditions, (a) the oil recovery curve; (b) the water saturation distribution inside the core plug after 40 days.