Oil Recovery by Spontaneous Imbibition from Weakly Water-Wet Rocks

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ABSTRACT

Spontaneous imbibition is of special importance to oil recovery from fractured reservoirs. Laboratory measurements of volume of liquid imbibed versus time are often used in the prediction of oil recovery. Imbibition measurements also provide a useful approach to the complex problem of characterizing the wetting properties of porous media. Correlation of a large body of data for imbibition of brine into porous media initially saturated with refined oil was achieved through a semi-empirical scaling group which includes permeability, porosity, interfacial tension, oil and brine viscosity, and the size, shape and boundary conditions of the sample.

The objective of the present study was to test this correlation for samples of different geometry for spontaneous imbibition under weakly water-wet conditions established by adsorption from crude oil with initial water saturation ranging from 14% to 31%. After establishing initial water saturations, the wettability of thirty-two core samples was changed to weakly water-wet by aging in an asphaltic crude oil at elevated temperature. Initial water saturation had a dominant effect on rate of oil recovery. Times for imbibition decreased by about 2 to 4 orders of magnitude with decrease in initial water saturation. Results for cores with the same initial water saturation but of different size and shape (cylindrical, annular, and rectangular) and boundary conditions (given by sealing off part of the rock surface with epoxy resin) were closely correlated. The presence of epoxy resin during aging in crude oil enhanced the decrease in water-wetness attained for cores and crude oil alone. The contribution to oil recovery by gravity segregation at very weakly water-wet conditions is addressed.

INTRODUCTION

Rate and extent of spontaneous imbibition provides a measure of wettability that is inherently dependent on surface energy. However, the mechanism of imbibition is highly complex and many factors besides wettability affect the rate of spontaneous imbibition and displacement efficiency. In previous studies, a semi-empirical scaling group was developed for recovery of oil from very strongly water-wet (VSWW) media (Ma et al., 1997). Dimensionless time, t_D, was defined by,

$$t_D = t_N \frac{\overline{k}}{f} \frac{s}{\sqrt{m_o m_w}} \frac{1}{L_c^2}$$
(1)

where t is the imbibition time; $\sqrt{k/f}$ is proportional to a microscopic pore radius; σ is the interfacial tension; $\sqrt{m_o m_w}$ is the geometric mean of the oil and brine viscosity. L_c is a characteristic length that compensates for sample size, shape and boundary conditions,

and is defined by $L_c = \sqrt{V/(\sum_{i=1}^n A_i / x_{A_i})}$, where V is the bulk volume of the core, A_i is the

open area of the *i*th face, and x_{Ai} is the distance from the no-flow boundary to the open face.

This scaling relationship has been shown to correlate imbibition data with variations in porosity, permeability, boundary conditions, and liquid viscosity ratios. The correlation was initially developed mainly for VSWW conditions with zero initial water saturation. Changes in dimensionless time for imbibition with change in water saturation at VSWW conditions have also been investigated (Viksund et al., 1998).

The applicability of the scaling group to wettability conditions other than VSWW needs to be investigated. Most oil reservoirs are considered to have some form of mixed wettability (Salathel, 1973). Mixed wettability arises because the distribution of connate water controls which parts of the rock surface are exposed to adsorption from crude oil. As the connate water saturation decreases, the exposed area increases and a larger change in wetting results. Change in the disjoining pressure of aqueous films with the increase in capillary pressure that accompanies decrease in water saturation may also be a factor in wettability alteration (Melrose, 1982; Kovscek et al., 1993).

Wettability change from VSWW conditions to mixed-wet can be induced by adsorption from crude oil (Jadhunandan and Morrow, 1991, 1995; Zhou et al., 1995, 1996). Changes in wettability can exhibit a wide spectrum of wetting behavior. The changes have been shown to depend on the crude oil type, the rock, aging time, temperature of aging, T_a , and of measurement, T_m , the brine composition and concentration, and the initial water saturation.

Experimental studies have shown that if a sandstone core is aged at ambient temperature, T_{a-amb} , decrease in imbibition rate measured at T_{m-amb} from very strongly water-wet is small. If the core is aged at elevated temperature, T_{a-high} , the imbibition rate at T_{m-amb} is reduced by up to several orders of magnitude relative to VSWW conditions. If the imbibition rate, for a core aged at T_{a-high} is measured at T_{m-high} , the imbibition rate is much higher than that measured at T_{m-amb} (Tang and Morrow, 1997). The controlling mechanisms for these differences are not well understood. In the present study the conditions T_{a-high} for aging and T_{m-amb} were chosen to provide weakly water-wet conditions that were readily distinguished from imbibition behavior for VSWW rocks. All conditions were held essentially constant except for initial water saturation, sample geometry, and boundary conditions.

EXPERIMENTAL

Cores: The rock samples in this study, 32 in all, were cut from the same block of Berea sandstone which had been tested for uniformity of properties. Different shapes of cores were prepared, including 1", 1.5" and 2" diameter cylindrical plugs of different lengths (some chosen to give cylinders of similar geometry), 2" outside diameter and 1" inside diameter annular plugs and rectangular blocks. Core dimensions and petrophysical properties are listed in Table 1. Some of the cores were partially coated with epoxy resin to obtain selected boundary conditions. The air permeabilities of the cores were all about 450 md, and the porosities were all close to 20%.

Brine: Synthetic sea water containing NaCl (28000 ppm), KCl (935 ppm), MgCl₂ (5365 ppm), CaCl₂ (1190 ppm), and 100 ppm of NaN₃ as biocide, was used as the brine phase. The total dissolved solids content was 35490 ppm. The interfacial tension between A95 crude oil and the synthetic seawater was 25.6 dynes/cm at room temperature measured by the du Nouy ring method.

Crude oil: An asphaltic crude oil, Alaska 95 (A95) from Prudhoe Bay, was used to change the wettability of the cores. The oil was degassed by vacuum treatment. The oil had 6.55 wt% of n-heptane asphaltenes and no detectable wax content. The viscosity of the evacuated oil was 40.3 cp at room temperature (22° C) and 18.5 cp at 55°C. The acid and base numbers were 0.24 and 2.2 respectively. The API gravity of the oil was 25.2. This oil was selected because it was known to induce significant change in wettability and would not give problems associated with wax deposition even at ambient temperature.

Core surfaces partially sealed with epoxy resin: Devcon 5 minute epoxy was used to partially seal the core surfaces to give different boundary conditions. The components are bisphenol (a diglycidyl ether resin), and a hardener that contained 2,4,6-tri (dimethylaminomethyl) phenol and polymercaptan curing agent. The partially sealed cores were set at room temperature to dry. The cores were then rinsed with tap water and dried at 110°C. The effect on core wettability of exposing the crude oil during aging to a block of the hardened epoxy resin was also tested.

Establishing initial water saturation: The core samples were first saturated with the synthetic sea water by vacuum. They were then left in the brine for about 10 days to attain ionic equilibrium. A porous plate apparatus (Soil moisture ceramic plate extractor) was used to establish the initial water saturation. Contact with the plate was ensured by seating in a very thin layer of fine cuttings obtained from Berea sandstone. Pressure differences of 10, 25, 40 and 55 psi, were applied sequentially (the capillary pressure in the core was always well below the applied pressure). Initial water saturations established by this procedure (after up to 10 days) ranged from 14-30% (see Table 1).

Saturation with crude oil: After establishing S_{wi} , the cores were saturated with A95 crude oil under vacuum. Any core which was determined, from mass balance, to be not fully saturated, was set under crude oil in a stainless steel aging cell at 800 psi for up to 3 days to ensure full saturation by liquids.

Aging: The cores containing initial water and crude oil were submerged in crude oil in aging cells which were then sealed. All of the cores were aged at 55°C for 10 days.

Imbibition test: After aging, the cores were set in glass imbibition cells filled initially with brine. All of the imbibition tests were performed at room temperature. Oil volume produced by imbibition of brine (expressed as percentage of original oil in place - %OOIP) versus time was recorded.

RESULTS AND DISCUSSION

Conditions for 32 imbibition tests are listed in Table 1 according to general classes of boundary conditions.

All-faces-opens cores (AFO):

Recovery by spontaneous imbibition versus time for weakly water-wet cores with all faces open are presented in Fig.1a. Imbibition time increased systematically over about three orders of magnitude with decrease in S_{wi} from 31.2% to 14.1%.

Table 1 Core sample dimensions and properties ($k_g \approx 450 \text{ md}, \phi \approx 20\%$)								
Core No.	L, cm	D, cm	d, cm*	φ, %	L _c	S _{wi} , %	Boundary conditions	
	or L ₁	or L ₂	or L ₃					
All faces op	en:							
S15-3	7.802	3.808	-	20.47	1.2727	31.2	AFO (all faces open)	
S2-3	5.884	5.03	-	19.91	1.5219	30.5	AFO	
S15-14	7.660	3.808	-	19.97	1.2701	27.6	AFO	
S15-5	7.812	3.808	-	20.24	1.2728	22.2	AFO	
S15-16	7.836	3.808	-	20.8	1.273	20.5	AFO	
S15-4	7.543	3.808	-	20.14	1.2680	20.3	AFO	
SA2-3	5.682	5.03	3.014	20.55	0.9500	19	AFO	
S2-8	4.721	5.03	-	20.27	1.4204	18.7	AFO	
S2-12	4.390	5.03	-	20.34	1.3818	18.1	AFO	
SS3**	L ₁ =3.22	L ₂ =4.49	L ₃ =5.44	18.84	1.1790	18	AFO	
SA2-4	4.293	5.03	3.014	19.84	0.9124	16	AFO	
S1-7	4.890	2.618	-	20.28	0.8656	15	AFO	
S2-14	3.612	5.03	-	20.39	1.267	14.1	AFO	
S2-10 [#]	4.520	5.03	-	20.67	1.3976	14.9	AFO	
Partially sea	led:							
S15-1	7.948	3.808	-	20.03	3.9740	27.1	TEO (two ends open)	
S2-11	3.744	5.03	-	20.3	1.0273	23.5	TEO	
S2-1	6.462	5.03	-	20.47	3.2310	23	TEO	
S15-18	6.023	3.808	-	20	3.0115	21	TEO	
S2-5	4.633	5.03	-	20.51	2.3165	21	TEO	
S1-5	4.599	2.622	-	19.7	2.2995	20.8	TEO	
S2-6	4.612	5.03	-	20.16	1.7784	20.4	TEC (two ends closed)	
S1-3	4.760	2.622	-	19.56	2.3800	19.7	TEO	
S15-6	7.451	3.808	-	20.89	1.3463	20.3	TEC	
SS1	L ₁ =4.30	L ₂ =3.00	L ₃ =5.42	19	2.7115	18.5	Two adjacent faces sealed	
							$L_1/L_3 \& L_2/L_3$	
S2-9	4.273	5.03	-	20.7	2.1365	18.2	TEO	
SS4	L ₁ =5.56	$L_2=4.41$	L ₃ =3.50	19.25	1.9534	18.1	TEO, both L_2/L_3 ends open	
S15-9	6.451	3.808	-	20.47	1.3463	17.8	TEC	
S1-4	4.837	2.618	-	19.9	0.9256	17.7	TEC	
S1-1	5.068	2.622	-	20.3	0.927	17.3	TEC	
S15-15	6.552	3.808	-	19.9	1.3463	17	TEC	
SA2-1	5.823	5.03	3.014	20.1	0.504	16.8	TEC	
S1-8	5.143	2.618	-	19.9	2.5715	16.8	TEO	

*inner diameter of the annular core

**rectangular core with dimensions L1, L2 and L3

[#]AFO core aged with epoxy resin in crude oil during aging

Results (Viksund et al, 1998) for recovery of mineral oil from Berea sandstone at very strongly water-wet (VSWW) conditions for S_{wi} ranging from 9.6% to 30% are included in Fig.1. Within this range of initial water saturation the imbibition times decrease with increase in S_{wi} but by much less than an order of magnitude. These results are shown as plots of recovery versus dimensionless time in Fig.1b. A general correlation for imbibition at VSWW conditions with zero initial water saturation is also

included in Fig.1b. This correlation is a fit of the model proposed by Aronofsky et al (1958) to a large body of imbibition data (Ma et al., 1997).

For chalk, scaled imbibition curves for $S_{wi} = 0$ were in close agreement with results for Berea sandstone, even though the permeability of chalk was over two orders of magnitude less than for sandstone, and the porosity was higher by a factor of about 2. Increase in S_{wi} in chalk from 0% to 34% resulted in increase in imbibition rate (Viksund et al., 1998), but, in contrast to Berea sandstone, scaled imbibition times were all less than for the correlation obtained for $S_{wi} = 0$. For both chalk and sandstone, the variation in imbibition time with initial water saturation at VSWW conditions was always less than an order of magnitude.

Scaled results for weakly water-wet conditions show a large systematic dependence on initial water saturation. The dominant effect of initial water saturation on the scaled imbibition data, especially for weakly water-wet conditions, requires that, for a meaningful test of scaling by L_c , the initial saturation should be essentially constant. Results for initial water saturation in the range 18.7 to 20.5% are shown in Fig. 2a. The agreement between results for duplicate tests with cores S15-4 and S15-16 demonstrates that close reproducibility can be obtained. The separation between these results and those for S2-8 and SA2-3 is greatly reduced after scaling by the characteristic length L_c (Fig. 2b). All other factors in the definition of t_D for the results presented in Fig.2b are very close to constant.

Partially sealed (PS) cores:

A data set for linear imbibition in cylindrical cores with two-ends-open (TEO) is shown in Fig.3a. Recovery vs. t_D is shown in Fig. 3b. As for AFO cores there is a distinct overall trend; scaled imbibition time increases with decrease in S_{wi} .

The same trend is observed for radial imbibition in cores with two-ends closed (TEC), and other boundary conditions (see Table 1) that involve partially sealed core surfaces (see Fig. 4a and 4b). Fig. 4 includes, in addition to data for TEC, results for an annular core with two ends closed, and two rectangular cores with two faces closed (see Table 1). As for the AFO cores, there was an overall trend of increase in imbibition time with decrease in S_{wi} . This is more systematic for the scaled results (cf. Figs. 4a and 4b).

As for the all faces open cores, a more definitive test of scaling is given by comparing results within a close saturation range. Data for cylindrical cores with either TEO or TEC in the saturation range of 19.7 to 21% are shown in Fig. 5a. The scaling group with L_c^2 ranging from 0.25 to about 16 gave close correlation of the results (Fig. 5b).

Results in the saturation range 17.3 to 18.5% were also tested (Fig. 6a). Although there was some degree of correlation it was less satisfactory for this data set (see Fig. 6b).

Effect of epoxy resin on wettability alteration:

The data points for the correlation of AFO cores shown in Fig. 2b with $S_{wi} = 18.7$ to 20.5% are included in Fig. 5b. Even though the average S_{wi} for the partially sealed (PS) core results is slightly higher than the AFO cores (20.35% vs. 19.6%), the relative values of t_D for the AFO cores are clearly less than for the PS cores.

This trend in relative values of t_D applies to all of the results. Scaled imbibition times for 50% recovery, $t_D(0.5)$, attained after about 12 weeks of imbibition, are plotted in Fig.7. Because of very slow continued imbibition, the end point saturations that were used to define $t_D(0.5)$ were not perfectly defined. However, the results for the AFO cores provide a well defined trend of increase in imbibition time, $t_D(0.5)$, with decrease in initial water saturation. The same plot for the PS cores show a comparable trend but with much more scatter; all of the values of $t_D(0.5)$ except for S1-8 lie above the trend line for AFO cores. The longer values of t_D for the PS cores show that, overall, the change in wettability was significantly larger than for the AFO cores. The difference provides a measure of the additional decrease in water-wetness resulting from the presence of the epoxy resin.

Use of the Devcon epoxy resin in experiments at ambient temperature with refined oil gave results for PS cores which could be correlated precisely with results for AFO cores (Zhang, et al., 1996). The results of the present work indicate that when cores with partially sealed faces were aged in crude oil, the presence of the epoxy resin caused the cores to undergo a larger change in wetting than cores with all faces open. The shift towards less water-wetness may be related to the presence of amine groups in the epoxy resin curing agent.

As a test of the effect of the epoxy resin on wetting, an AFO core was prepared with a block of solidified resin in the crude oil during aging. Unlike the partially sealed cores, the resin was not in direct contact with the core. The initial water saturation was 14.9%. After aging for 10 days at 55°C, the imbibition rate of the AFO core was consistent with that of the PS cores. Imbibition results are included in Fig. 7 as an insert. These results confirmed that interaction between the resin and the crude oil caused the PS cores to become less water-wet than the cores exposed to crude oil alone. This could be avoided in future work by, for example, sealing the core with low melting point metal or the use of boundary surfaces such as PTFE heat shrink tubing. However, in practice only AFO cores are used in the majority of imbibition tests. The results obtained for PS cores with epoxy resin are still of interest with respect to the problem of scaling.

Inspection of the scaled results in Fig. 5b and the $t_D(0.5)$ values in Fig. 7 shows that in four cases they are in close agreement. The high and low values of $t_D(0.5)$, for S15-6 and S1-3 respectively, result from differences in extent of imbibition rather than in curve shape (see Fig. 5b).

The values of $t_D(0.5)$ for the PS cores in the saturation range 17.3 to 18.5% show much more scatter (Fig. 7) as indicated by the range of deviation from the trend for AFO cores. The scatter is related to the spread in the scaled results shown in Fig. 6b. Bearing in mind the difference in water saturations, the fit to the data for AFO cores and the results for PS cores shown in Fig. 5b appear to span the extremes of the scaled data shown in Fig. 6b.

Two key factors in characterizing the imbibition process are the imbibition time, for example the time required for 50% of final recovery $t_D(0.5)$, and the fraction of in place oil recovered, otherwise known as the microscopic displacement efficiency. It is unlikely that a general account can be developed of change in imbibition behavior with wettability because of the number of variables that are involved, their individual complexity, and the complexity of their interactions. Zhou et al.(1996) reported imbibition results for water saturations of 15%, 20% and 25%. In these tests, the final recovery showed greater sensitivity to initial water saturation and the imbibition time showed less sensitivity than found in the present work. However, there were many differences in the two studies

including the crude oil and an aging temperature of 85°C, compared to aging at 55°C in the present work.

Scaling of wettability and gravity

The values of $t_D(0.5)$ for AFO cores shown in Fig.7 were normalized with respect to $t_D(0.5)$ (= 14.5) for VSWW with $S_{wi} = 0$, to obtain the scaling factor presented in Fig.8. Normalized values of $t_D(0.5)$ for VSWW conditions with an initial water saturation are also presented. The increase in $t_D(0.5)$ caused by change in wetting is given by the ratio of scaling factor for the weakly water-wet and strongly water-wet conditions. For example at 20% S_{wi} , the imbibition time resulting from interaction with crude oil is increased by $\Delta t_D(0.5) = 514$ (see Fig.8).

A condition in the development of scaling laws for imbibition was that the effect of gravity be neglected (Rapopport, 1955; Mattax and Kyte, 1962). If capillary forces are sufficiently small, gravity segregation will make a significant contribution to oil recovery. If gravity is included, the expression for dimensionless time for linear imbibition will have the form

$$t_D(c+g) = t \frac{k/f}{L_c^2 \sqrt{\boldsymbol{m}_w \boldsymbol{m}_o}} (P_{ci} f(\Theta) + \frac{\Delta \boldsymbol{r} g L_c^2}{L_H})$$
(2)

where $t_D(c+g)$ is the dimensionless time for imbibition that includes capillary and gravity forces, P_{ci} is a representative imbibition capillary pressure and is proportional to

$$\frac{S}{\sqrt{k/f}}$$
, $f(\Theta)$ is a wettability factor, and L_H is the vertical height of the sample.

For a vertical Berea sandstone core of length 7 cm, with difference in density of oil and brine of 0.1g/ml, the buoyancy force that drives gravity segregation is given by $\Delta \mathbf{r}gL_H = 29 \, dynes / cm^2$.

A representative capillary pressure in Berea sandstone at strongly water-wet conditions is about 23×10^3 dynes/cm². If wettability change reduces the pressure by about three orders of magnitude, the relative contribution to oil recovery by gravity forces will become significant.

If capillary forces become negative and exceed gravity, imbibition will not occur. This condition, which corresponds to an Amott wettability index (Amott, 1959) to water of zero, was not observed in the present work. (Imbibition behavior at water saturations lower that 15% still need to be tested.) It appears that imbibition is promoted by slow change towards increased water-wetness during the course of the test. Such evidence of slow dynamic changes towards increased water-wetness further complicates interpretation of results with respect to the relative role of surface and gravity forces when imbibition is very slow.

Aronofsky model of imbibition behavior for change in S_{wi} :

The Aronofsky model (Aronofsky et al, 1958) provides a reasonable one parameter fit for VSWW cores (see Fig. 1b)

$$\frac{R}{R_{\infty}} = 1 - e^{-at_D} \tag{3}$$

where R is the oil recovery at time t_D , R_{∞} is the final recovery, and a = 0.05.

The oil recovery at $t_D(0.5)$, R(0.5) was used to determine the constant *a* for saturations of 15 to 30% (Fig.9), where $a = -\ln[1 - R(0.5)]/t_D(0.5)$. Although this model does not give a close fit to imbibition curve shape for the weakly water-wet conditions, it does provide a clear illustration of the dominant effect of initial water saturation on oil recovery by spontaneous imbibition.

CONCLUSIONS

The following conclusions apply to the conditions of generation of mix wettability used in the present work. Care must be taken in applying results beyond the range of the experimental data.

- 1. Initial water saturation has a dominant influence on the rate of oil recovery.
- 2. A characteristic length used to scaled the effect of sample size, shape, and boundary conditions for strongly water-wet cases gave satisfactory scaling for weakly water-wet conditions.
- 3. Scaled imbibition time decreased systematically with decrease in initial water saturation.
- 4. Aging of sandstone with crude oil in the presence of epoxy resin can cause a significant shift towards reduced water-wetness relative to that induced by crude oil alone.

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NOMENTCLATURE

a – constant for Aronofsky model,	t _D – dimensionless imbibition time,			
A_i – open area of ith face of the sample,	$t_D(c+g)$ – dimensionless imbibition time			
cm^2 ,	that includes capillary and gravity			
$f(\Theta)$ - wettability factor,	forces, x _{Ai}			
k – gas permeability, md,				
L _c – characterize length, cm,				
L_H – vertical height of the sample, cm	φ - porosity, %			
P _{ci} – imbibition capillary pressure,	$\Delta \rho$ - oil/water density difference, g/ml.			
R – oil recovery by imbibition, OOIP%,	σ - oil-water interfacial tension,			
R_{∞} - final oil recovery by imbibition,	dynes/cm.			
OOIP%,	$\mu_{\rm w}$ – water viscosity, cp,			
t – imbibition time, min,	μ_{o} – oil viscosity, cp.			

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Fig.2 Application of the characteristic length to scaling imbibition data for weakly water-wet cores with all faces open.



Fig. 3 Effect of initial water saturation on imbibition rate and oil recovery for weakly water-wet cores with two ends open.



Fig.4 Effect of initial water saturation on imbibition behavior for weakly water-wet cores with partially sealed surfaces.



(a) Imbibition curves for S_{Wi} = 19.7 \sim 21%

r (00IP%)

(b) Correlated results for PS cores

Fig. 5 Scaling of imbibition data for weakly water-wet cores with partially sealed surfaces and initial water saturation in the range 19.7 \sim 21%.



(a) Recovery vs. time



Fig.6 Scaling of results for weakly water-wet cores with partially sealed surfaces and with initial water saturation in the range 17 to 18.5%.



Fig.7 The relationship between S_{wi} and imbibition time for 50% oil recovery. Insert shows effect of epoxy resin on imbibition by AFO cores.



Fig.9 Aronofsky model based on values of $t_{\rm D}$ at 50% recovery for AFO cores.



Fig.8 Effect of initial water saturation and change in wettability on imbibition time for 50% recovery (relative to VSWW with $S_{wi} = 0$).