EFFECT OF SALINITY ON OIL RECOVERY
BY SPONTANEOUS IMBIBITION

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ABSTRACT

Companion sets of waterflood and spontaneous imbibition data for duplicate core plugs provide indication of the changes in wettability and microscopic displacement efficiency that accompany reduction in salinity of the invading phase. However, the effect of salinity on waterflood recovery for reservoir rocks is often investigated without obtaining diagnostic information on wettability, mainly because suitable duplicate core plugs are not available. In the present work, emphasis is placed on the effect of salinity on oil recovery by spontaneous imbibition. Imbibition data was obtained for outcrop sandstone, limestone, and reservoir sandstone. In all, the results spanned differences in porosity, permeability, pore structure, mineralogy, initial water saturation, and initial and invading water composition. Two crude oils of moderate viscosity (>50 cP at ambient temperature) and high asphaltene content were used in all imbibition tests, apart from reference tests made on recovery of refined oil at very strongly water-wet (VSWW) conditions. In addition to oil recovery versus time, changes in pH and conductivity of the effluent brine were monitored and the produced crude oil was checked for the presence of water-in-oil emulsions. Use of low salinity brine at the outset of an imbibition test resulted in higher microscopic displacement efficiency relative to imbibition of high salinity brine for both outcrop and reservoir sandstone samples. Rates of imbibition were comparable. When recovery by invasion of high salinity brine had essentially ceased, in many cases, change of the invading brine to low salinity resulted in additional oil production. For the reservoir sandstone, after completion of high and low salinity imbibition, additional oil recovery was obtained by low salinity waterflooding.

INTRODUCTION

Jadhunandan and Morrow (1991) showed that brines with varied ratio and amount of mono to divalent ions, can have significant effect on the rate and extent of oil recovery by spontaneous imbibition. It was also reported that waterflood recoveries increased as water wetness, assessed by a form of Amott wettability index, decreased towards neutral wettability (Jadhunandan and Morrow, 1991). Several other subsequently measured companion waterflood and spontaneous imbibition data sets also showed increase in waterflood recovery with decrease in water wetness (Zhou et al. 2000; Tie and Morrow, 2005).

Investigation of the effect of brine concentration on oil recovery often showed significant increase in laboratory waterflood recoveries with decrease in salinity for duplicate outcrop core plugs. However, companion data sets for waterfloods and spontaneous imbibition both showed increased recovery with decrease in salinity (Tang and Morrow, 1999a, 1999b, 2002; Zhang and Morrow, 2006). No consistent explanation has yet been developed of the effect of salinity on oil recovery. Tang and Morrow (1999a) concluded that adsorption from crude oil, and the presence of clay and initial water saturation are necessary conditions for increased oil recovery through reduced salinity. It was later shown that both adsorption from crude oil and the presence of the crude oil/brine interface were necessary (Zhang et al., 2007). In spite of widespread investigation of low salinity waterflooding, the sufficient conditions and flow mechanisms for increased oil recovery have yet to be clearly identified. Thus, when field application is under consideration, each specific reservoir system should be tested for low salinity response.

However, the amount and quality of available reservoir core is usually limited, and heterogeneity may preclude the possibility of developing companion waterflood and imbibition tests for comparable core material. To date, most evaluation studies have been based on laboratory waterfloods. Spontaneous imbibition measurements provide an alternative approach which is simpler and less costly than laboratory waterfloods, and also provides more definitive indication of wettability variations associated with changes in salinity. For example, Ligthelm et al. (2009) showed that improved oil recovery by imbibition was obtained for a Middle Eastern microcrystalline limestone when the aquifer brine was modified to include a higher sulfate to calcium ratio. However, the low salinity aquifer brine itself did not give improved oil recovery by spontaneous imbibition for the particular limestone (Ligthelm et al., 2009).

Tang and Morrow (1999b) observed increases in imbibition oil recovery of 8 to 13% of original oil in place (OOIP) for Berea sandstone with reservoir crude oil, synthetic reservoir brine as the connate water, and immersion in five different diluted brines. Zhang and Morrow (2006) and Zhang et al. (2007) also observed increase in imbibition oil recovery with decrease in salinity.

In the present work, the effects of salinity on oil recovery are reported for 27 imbibition tests on outcrop oolitic limestone and Berea sandstone, and a reservoir sandstone that had high siderite (iron carbonate) content. Omission of the usual extended period of aging a core in crude oil prior to commencement of tests on oil recovery was also investigated.

Scaling of imbibition data
In assessment of wettability, imbibition data is often compared using a dimensionless time defined by Ma et al. (1997),

$$t_{D,Ma} = t \sqrt{\frac{k}{\phi}} \frac{\sigma}{\sqrt{\mu_w \mu_{nw}}} \frac{1}{L^2_c}$$

where t_D is the dimensionless time, t is the imbibition time, k is the permeability of the rock, ϕ is the porosity of the rock, σ is the interfacial tension between the oil and brine phases, μ_w is the brine viscosity, μ_{nw} is the oil viscosity, and L_c is the characteristic length (Ma et al., 1997).

Fischer et al. (2008) expanded the range of variation in viscosity ratio to more than four orders of magnitude by use of brine-glycerol mixtures to increase the viscosity range of the aqueous phase. The extended data was satisfactorily correlated by changing the empirical correlation of the effect of the aqueous and oleic phase viscosities by the scaling group of Mason et al. (2010)

$$t_{D,MFMR} = t\sqrt{\frac{k}{\phi}} \frac{\sigma}{\mu_w\left(1 + \sqrt{\mu_{nw}/\mu_w}\right)} \frac{2}{L_c^2}$$

The main difference between the modified scaling group above and the Ma et al. (1997) scaling group is the viscosity term, $\mu_w + (\mu_w\mu_{nw})^{1/2}$ instead of $(\mu_w\mu_{nw})^{1/2}$. The two scaling groups are empirical with respect to viscosity ratio. Both give close correlation of results for increase in oil phase viscosity with brine as the aqueous phase. In this paper, the modified scaling group of Mason et al. (2010) is used to assess changes in imbibition behavior that result from change to mixed-wet conditions.

EXPERIMENTAL METHODS
Cores
Edwards Garden City (GC) Limestone
Edwards GC is an oolitic grainstone (Figure 1(a)) which consists of spherical ooids and other fossil fragments cemented by calcite (Tie, 2006). The limestone was quarried from the Edwards formation near Garden City, Texas, and supplied as 12x12x6 inch blocks. 11 cores of 1.5 inches diameter were cut, oven dried at 100ºC, and then placed in a desiccator at ambient conditions. The gas permeabilities ranged from about 7 to 34 md. The porosities ranged from about 19 to 22% (Table 1). The BET surface area by N_2 adsorption and Cation Exchange Capacity (CEC) of Edwards GC limestone was 0.20 m^2/g and 0.26 x 10^3 meq/g respectively (Tie, 2006).

Berea Sandstone
Berea sandstone cores of 1.5 inches diameter were cored from blocks supplied by Cleveland Quarries, Ohio. This rock type mainly consists of quartz, feldspar, and lithic fragments with small amounts of sparry dolomite cement, kaolinite, and chlorite (Figure 1(b)). The BET surface area was about 0.65 m^2/g for the Berea sandstone (Tie, 2006). The gas permeabilities ranged from 555 to 639 md; the porosities were about 21% (Table 1).

RS Reservoir Sandstone
RS reservoir cores mainly consist of quartz. Some samples were high in siderite content which may tend to make the rock less water-wet. There were also considerable amounts of feldspar. The total clay content was high in some samples, with glauconite being

predominant. Micro-porosity is associated with glauconite, feldspar, and other clays and lithic fragments (Figure 1(c)). The cores were cleaned by soxhlet extraction for several months before being selected for experiments. The gas permeabilities and porosities covered a wide range (Table 2). Companion plugs were available for comparison of imbibition tests. CEC values ranged from about 5 to 9 meq/g (Table 2).

Oleic Phases
Two mineral oils, Soltrol 220 with a viscosity of 3.8 cp (Low Viscosity Oil – LVO) and white mineral oil with a viscosity of 173.6 cp (High Viscosity Oil – HVO) were used for the VSWW experiments with Edwards GC limestone. The mineral oils were first contacted with activated alumina and silica gel to remove polar contaminants and then filtered. They were vacuumed for three to four hours to ensure removal of dissolved gas. An Alaskan crude oil (WP) was used in tests on Mixed-Wet (MXW) Edwards GC limestone and Berea sandstone and RS crude oil was used for the RS reservoir sandstone experiments. The crude oils were filtered and vacuumed before use. The asphaltene contents of the two crude oils are comparable, but the WP crude oil has higher viscosity than the RS crude oil. Properties of the oils are given in Table 3.

Aqueous Phases
A synthetic (SC) seawater with 35.5875 g/ L of Total Dissolved Solids (TDS) was used for the experiments on VSWW and MXW Edwards GC (Table 4). NaN_3 was added to the synthetic seawater to prevent bacterial growth. Field water analyses were used as a basis for preparation of the RS connate brine and the synthetic RS seawater which was used as the invading brine in experiments on RS reservoir sandstone and Berea sandstone. The RS low salinity brine composition was based on water analysis of an aquifer local to the reservoir (Table 4).

Core Preparation
Confining pressures of 300 and 500 psi were used during measurements of nitrogen gas permeabilities. Porosity, obtained by helium porosimetry, agreed with porosity determined by the weight difference of a dry versus fully saturated sample. VSWW Edwards GC cores with no initial water saturation were prepared by vacuum saturating the cores with a mineral oil followed by pressure saturating at 1000 psi. The Edwards GC and Berea MXW cores were first vacuum saturated in SC seawater and pressurized at 1000 psi for about a day to ensure full saturation. Cores were then left for at least ten days to reach ionic equilibrium at ambient temperature and pressure. The initial water saturation was established by use of a porous plate. The cores were then vacuum saturated and pressurized in the crude oil and aged at 75ºC (T_a) for ten days. A porous plate was used to establish initial water because this method has the ability to produce similar initial water saturations within the same rock type, and also to achieve low initial water saturations in the relatively low permeability Edwards GC cores. Comparable initial water saturations within each set of cores facilitate comparison of the effect of differences in invading brine salinities. The weight of the cores was recorded for the porosity and mass balance calculations. RS reservoir sandstone cores were saturated with

the RS connate brine and the water content was reduced to initial values by centrifuging at a speed equivalent to about 250 psi capillary pressure. The usual laboratory methods of centrifuging and flow of oil to establish initial water saturation resulted in comparatively high initial water saturations for the RS cores. Cores were then oil flooded with RS crude oil. A check on mass balance was made during each core preparation step to ensure full saturation of the core. These RS samples were not aged in RS crude oil or RS connate brine partly due to time limitations, but also to test the effect of low salinity imbibition for nominal aging times of zero. The prepared cores were placed in glass imbibition cells and immersed in the desired brine. All of the VSWW experiments were performed at ambient conditions and the MXW experiments were performed at 60ºC (T_m). The oil recovery versus time was recorded until the imbibition had stabilized or the imbibition rate was extremely slow. Four of the RS reservoir sandstone samples were waterflooded with RS low salinity brine at the end of imbibition experiments.

RESULTS AND DISCUSSION

VSWW Edwards GC Limestone
The VSWW Edwards GC experiments with either 3.8 cp mineral oil or 173.6 cp mineral oil did not show any trend of increase in oil recovery with decrease in salinity (Figure 2). The VSWW residual oil saturations obtained by spontaneous imbibition were lower than the waterflood residual oil saturations reported for VSWW Edwards GC limestone by Tie and Morrow (2005).

MXW Edwards GC Limestone
For the Edwards GC, imbibition tests with SC seawater and 1/10, 1/20, 1/50, and 1/100 dilution of SC seawater crude-oil production, as for the tests with refined oil, did not show a systematic trend of increase in recovery with decrease in salinity (Figure 2). The initial water saturations are comparable in this set of experiments. The MXW imbibition residual oil saturations were similar to those for recovery of refined oil by VSWW imbibition whereas Tie and Morrow (2005) found that waterflood residuals for MXW Edwards GC limestone were much lower than for VSWW conditions. Possible causes are differences in the mechanism of spontaneous versus forced imbibition and other factors such as sample heterogeneity.

Berea Sandstone
For mixed wettability Berea sandstone, recovery by imbibition of RS low salinity brine produced, on average, about 8% OOIP more than for imbibition of RS seawater (Figure 3). The initial water saturations for this data set were comparable as well. The slopes of the imbibition curves, which reflect wettability through the capillary driving force, did not vary much even though there was an increase in final oil recovery by imbibition of RS low salinity brine.

RS Reservoir Sandstone
RS 5a and RS 5b showed good reproducibility of results for immersion in RS seawater regardless of differences in initial water saturations (Figure 4). RS 6a and RS 6b differed

in gas permeabilities in addition to initial water contents (Figure 4), but once the data were correlated with the Mason et al. (2010) scaling group, the results for those two cores showed satisfactory agreement.

Imbibition rates were faster and the final oil recoveries were higher in both sets of experiments for which cores were first immersed in RS low salinity brine to give recovery of 30.5% OOIP for RS 6a and 16.7% for RS 4 (Figure 5). The faster imbibition rate for RS 6a might be due to its higher initial water saturation compared to that of RS 4 (47.7% vs. 26.0%). RS 6b, which is a duplicate core plug of RS 6a, gave a comparable final oil recovery (27.3% OOIP). However, RS 2 showed a lower final oil recovery (19.0% OOIP) when immersed in low salinity water even though RS 6b and RS 2 had similar initial water contents. RS 1, RS 5b, RS 3, and RS 5a were immersed in RS seawater and produced 7.5%, 6.2%, 12.7%, and 9.3% OOIP respectively. RS 3 and RS 5a produced more oil, probably due to their higher initial water saturations. Nevertheless, RS 1 and RS 5b did not show much difference in final oil production despite the differences in initial water saturation.

The immersed brine phase was changed in the second set of experiments to further evaluate the effects of salinity and composition on oil recovery by what will be referred to as stage 2 imbibition. The two cores, which were transferred from RS seawater to RS low salinity brine, produced additional oil. The two cores transferred from low salinity water to RS seawater did not produce additional oil (Figure 6). The crude oil and brine used for each experimental together with the initial water saturations of the cores and final oil recoveries are summarized in Table 5.

The pH of the brines as prepared decreased in all batch-type imbibition experiments showed. The conductivity of the brine decreased for post-imbibition of RS seawater, but increased for post-imbibition of RS low salinity brine. The final oil recovery versus the total clay content showed a positive linear relationship for stage 1 RS seawater imbibition and a negative linear relationship for stage 1 RS low salinity water imbibition. The final oil recovery after the post-imbibition waterflood showed a positive linear relationship with respect to the measured cation exchange capacity (CEC).

CONCLUSIONS
Neither VSWW Edwards GC experiments nor MXW Edwards GC experiments showed an increase in oil recovery when invading brine salinity was decreased. Variation in brine salinity and composition showed improved oil recovery by imbibition for both Berea sandstone and RS reservoir sandstone. Berea sandstone did not show much difference in slope the imbibition curves (only slight increases in the rate and final oil recoveries). But, the RS reservoir cores did show slope differences and imbibition rate changes for immersion in RS low salinity brine against RS seawater. The duplicate RS core samples were scaled satisfactorily by the Mason et al. (2010) scaling group. Brine pH after imbibition decreased in all of the RS reservoir core experiments. Brine conductivity after imbibition decreased for RS seawater imbibition and increased for low salinity water imbibition. While a strong correlation has not been established for final oil recoveries (%

OOIP) versus mineral contents, there was a satisfactory correlation of final oil recovery with total clay content.

ACKNOWLEDGEMENTS

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Table 1. Edwards GC and Berea core properties

Core	D (cm)	L (cm)	φ (%)	k_g (mD)
T2WTC8a	3.75	7.62	21.7	15.1
T2WTC8b	3.75	7.74	20.5	12.2
T2WTC9a	3.75	7.63	21.8	20.9
T2WTC11a	3.75	7.98	19.0	10.5
T2WTC11b	4.70	7.53	18.9	6.6
T2WTC12a	3.75	7.68	20.2	12.4
EGC1	3.80	6.91	21.0	32.8
EGC2	3.80	6.78	21.4	34.5
EGC3	3.80	6.70	20.3	27.4
EGC5	3.80	6.65	21.3	34.3
EGC11	3.79	6.91	20.2	30.9
BS1	3.79	7.82	21.1	604.0
BS3	3.79	7.90	21.3	638.9
BS7	3.79	8.01	21.0	555.0
BS10	3.79	7.03	21.4	625.3

Table 2. RS core properties

Core	D (cm)	L (cm)	φ (%)	k_g (mD)	CEC (meq/g)	Glauconite (%)	Siderite (%)	Total clay (%)
RS 1	3.78	5.19	26.4	49.4	7.12	3.1	25.9	22.4
RS 5b	3.79	4.25	24.8	89.2	5.47	1.8	40.4	19.6
RS 6a	3.79	5.61	26.3	74.0	5.12	2.7	11.5	9.3
RS 4	3.78	5.42	24.7	12.4	6.91	5.0	40.9	17.7
RS 3	3.77	4.76	32.0	226.3	9.15	4.7	30.2	31.8
RS 5a	3.79	5.34	20.8	73.0	5.47	1.8	40.4	19.6
RS 6b	3.80	3.34	26.9	163.7	5.12	2.7	11.5	9.3
RS 2	3.78	5.43	24.6	52.4	5.55	2.7	3.3	12.5

Table 3. Crude oil and mineral oil properties

Oleic phase	Viscosity μ (cp)	Density ρ (g/cm³)	Interfacial tension $\sigma_{Oil/Brine}$ (dyne/cm) (22 ^{0}C)	Asphaltenes (%)	Acid # (mg KOH/ g oil)	Base # (mg KOH/ g oil)
RS	97.4 (22ºC) 18.8 (60ºC)	0.9228 (22ºC) 0.8988 (60ºC)	25.0 (RS Seawater) 27.5 (RS Low Sal) 24.6 (RS Connate)	6.61 (n-C_6)		
WP	111.2 (22ºC) 20.1 (60ºC)	0.9125 (22ºC) 0.9070 (60ºC)	19.5 (SC Seawater)	6.30 (n-C_6)	1.46	2.49
LVO	3.8	0.7820	52.7 (SC Seawater)			
HVO	173.6	0.8720	54.4 (SC Seawater)			

Table 4. Compositions of RS seawater, RS low salinity brine, RS connate brine, and
SC seawater solutions

Component	RS seawater g/L	RS low salinity g/L	RS connate g/L	*SC seawater g/L
NaCl	22.8443	2.0328	22.855	28.0000
KCl	0.6678	0.0125	0.0993	0.9350
$MgCl_2.6H_2O$	9.7888	0.0628	0.0921	5.3625
$CaCl_2.2H_2O$	1.3279	0.1922	0.1908	1.1900
$NaHCO_3$	0.2051	0.5186	0.055	
Na_2SO_4	3.5920			
TDS	38.4259	2.8189	23.2922	35.4875

* 0.1 g/L of NaN_3 was added as a biocide

Table 5. Oil, brine, initial water saturation, and final oil recovery

Core	Oil	Brine	S_{wi} (%)	R_f (% OOIP)
Stage 1 imbibition				
T2WTC8a	LVO	SC seawater	0	43.6
T2WTC8b	LVO	1/10 SC seawater	0	42.2
T2WTC9a	LVO	1/100 SC seawater	0	42.5
T2WTC11a	HVO	SC seawater	0	39.6
T2WTC11b	HVO	1/10 SC seawater	0	40.0
T2WTC12a	HVO	1/100 SC seawater	0	40.0
EGC1	WP	SC seawater	8.2	42.6
EGC2	WP	1/10 SC seawater	6.6	40.6
EGC3	WP	1/20 SC seawater	6.6	39.0
EGC5	WP	1/50 SC seawater	7.3	43.9
EGC11	WP	1/100 SC seawater	6.5	42.2
BS1	WP	RS low sal	18.3	56.2
BS3	WP	RS low sal	18.9	57.2
BS7	WP	RS seawater	17.4	47.6
BS10	WP	RS seawater	17.5	49.1
RS 1	RS	RS seawater	34.9	7.5
RS 5b	RS	RS seawater	42.4	6.2
RS 6a	RS	RS low sal	47.7	30.5
RS 4	RS	RS low sal	26.0	16.7
RS 3	RS	RS seawater	52.6	12.7
RS 5a	RS	RS seawater	52.0	9.3
RS 6b	RS	RS low sal	31.2	27.3
RS 2	RS	RS low sal	33.0	19.0
Stage 2 imbibition				
RS 3	RS	RS low sal	52.6	31.0
RS 5a	RS	RS low sal	52.0	30.6
RS 6b	RS	RS seawater	31.2	27.3
RS 2	RS	RS seawater	33.0	19.0
Post-Imbibition				Waterflood
RS 3	RS	RS low sal	52.6	68.6
RS 5a	RS	RS low sal	52.0	34.4
RS 6b	RS	RS low sal	31.2	37.9
RS 2	RS	RS low sal	33.0	47.0

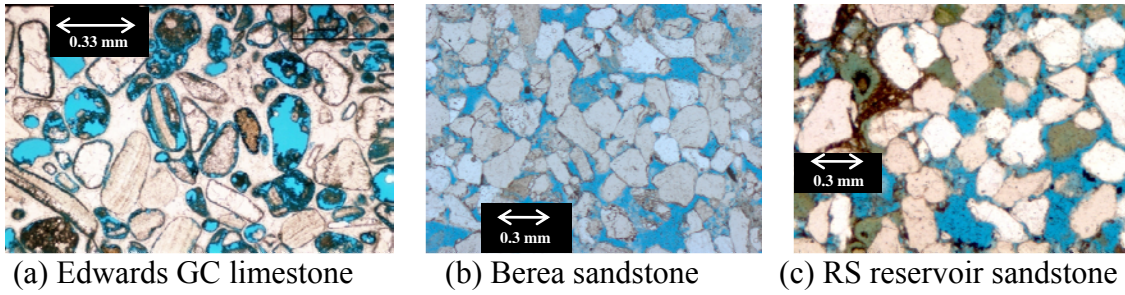(a) Edwards GC limestone   (b) Berea sandstone   (c) RS reservoir sandstone

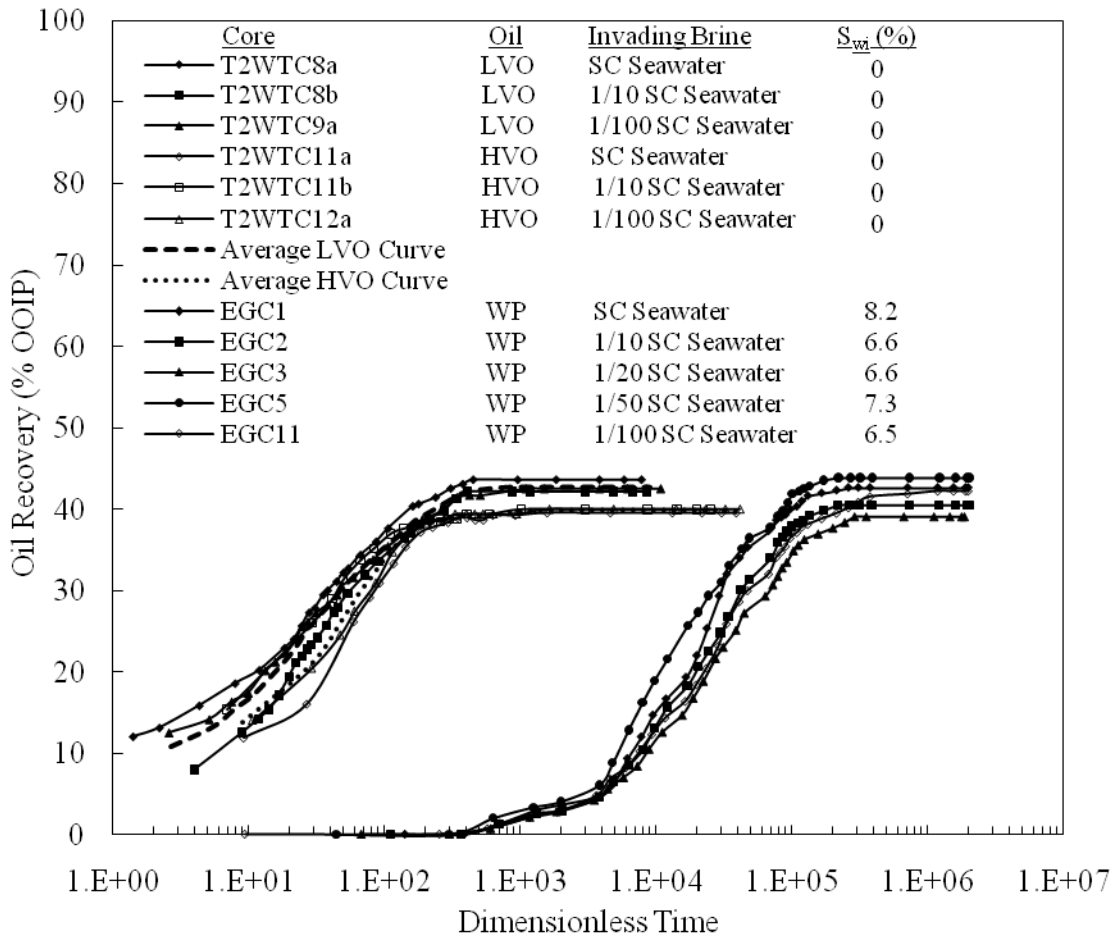Figure 1. Thin sections of outcrop and RS reservoir core samples

Figure 2. Recovery of WP crude oil (cores aged for 10 days at S_{wi} at 75°C and tested at 60°C) and low and high viscosity mineral oil, by spontaneous imbibition of SC seawater and diluted brines into Edwards GC limestone
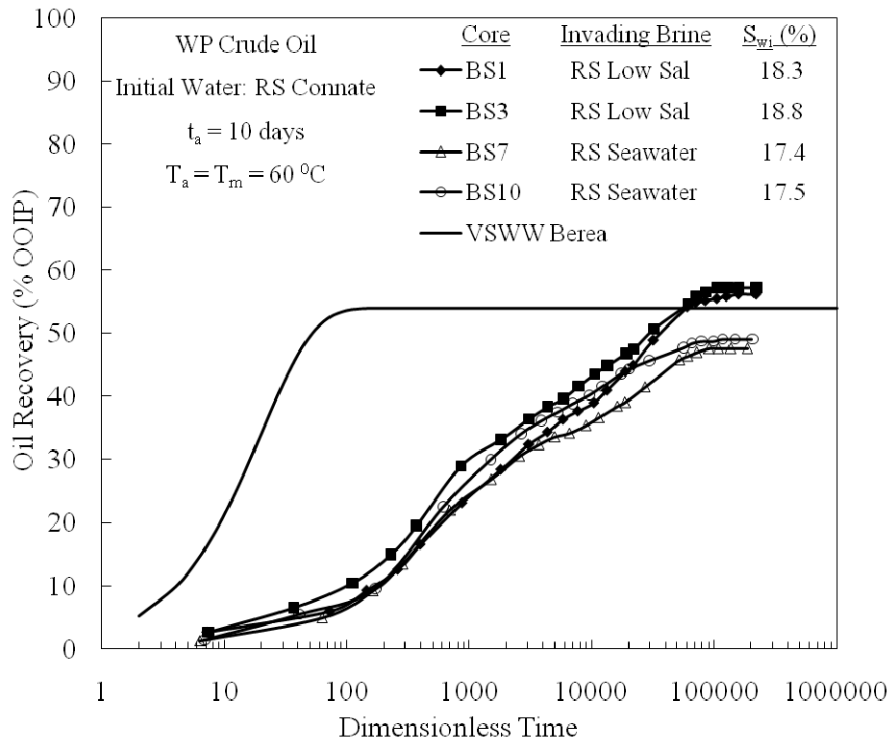
Figure 3. Recovery of WP crude oil by spontaneous imbibition of RS seawater and RS
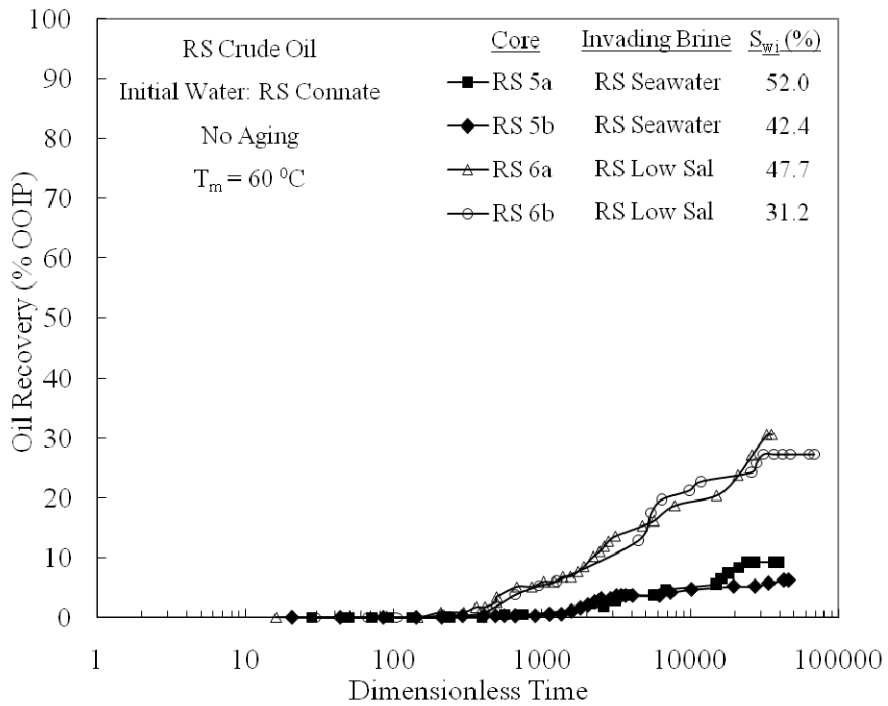   low salinity brine into Berea sandstone

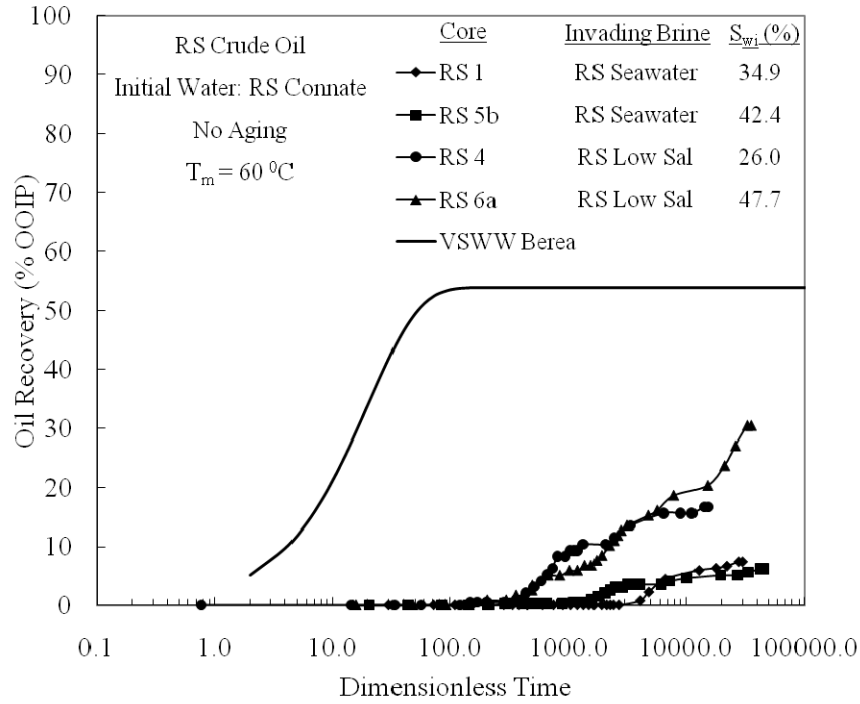Figure 4. Reproducibility of duplicate RS reservoir cores

Figure 5. Comparison of recovery of RS crude oil by spontaneous imbibition of RS
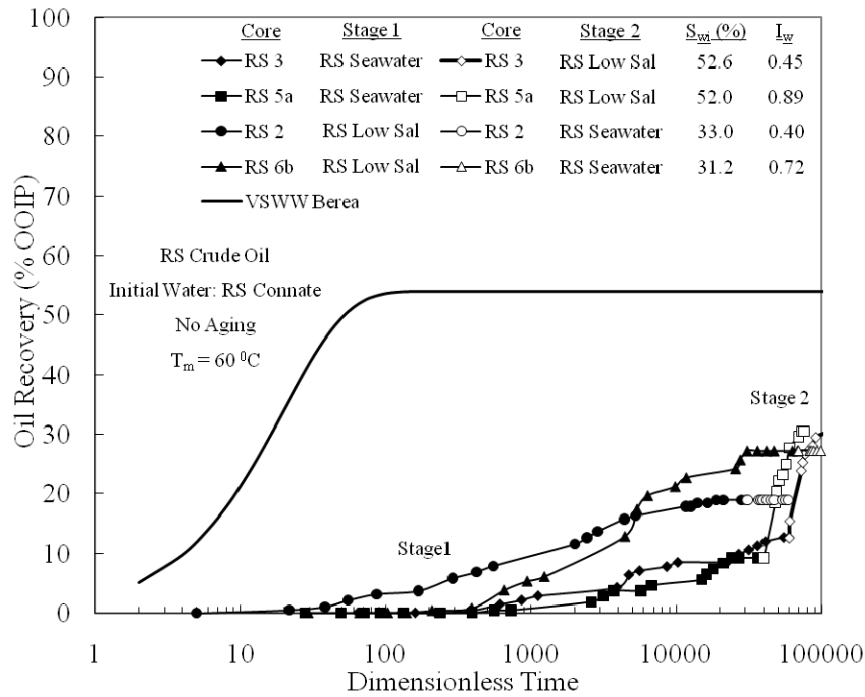   seawater and low salinity brine

Figure 6. Recovery of RS crude oil by spontaneous imbibition of RS seawater followed
          by low salinity brine and low salinity brine followed by RS seawater.