

Article

# Study of the Effect of Clay Particles on Low Salinity Water Injection in Sandstone Reservoirs

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**Abstract:** The need for optimal recovery of crude oil from sandstone and carbonate reservoirs around the world has never been greater for the petroleum industry. Water-flooding has been applied to the supplement primary depletion process or as a separate secondary recovery method. Low salinity water injection is a relatively new method that involves injecting low salinity brines at high pressure similar to conventional water-flooding techniques, in order to recover crude oil. The effectiveness of low salinity water injection in sandstone reservoirs depends on a number of parameters such as reservoir temperature, pressure, type of clay particle and salinity of injected brine. Clay particles present on reservoir rock surfaces adsorb polar components of oil and modify wettability of sandstone rocks to the oil-wet state, which is accountable for the reduced recovery rates by conventional water-flooding. The extent of wettability alteration caused by three low salinity brines on oil-wet sandstone samples containing varying clay content (15% or 30%) and type of clay (kaolinite/montmorillonite) were analyzed in the laboratory experiment. Contact angles of mica powder and clay mixture (kaolinite/montmorillonite) modified with crude oil were measured before and after injection with three low salinity sodium chloride brines. The effect of temperature was also analyzed for each sample. The results of the experiment indicate that samples with kaolinite clay tend to produce higher contact angles than samples with montmorillonite clay when modified with crude oil. The highest degree or extent of wettability alteration from oil-wet to intermediate-wet state upon injection with low salinity brines was observed for samples injected with brine having salinity concentration of 2000 ppm. The increase in temperature tends to produce contact angles values lying in the higher end of the intermediate-wet range (75°–115°) for samples treated at 50 °C, while their corresponding samples treated at 25 °C produced contact angle values lying in the lower end of intermediate-wet range.

**Keywords:** clay particles; wettability; temperature's effect; contact angle; sandstone reservoirs

## 1. Introduction

Low salinity water flooding (LSWF) is one such modified application of traditional water flooding techniques. Field tests and core-flooding experiments have verified that LSWF improves the oil recovery process by 5%–38% more than that by traditional water flooding techniques [1–4]. The usage of brines with low salinity concentration and lower ionic strength plays a significant role in the wettability alteration in both sandstone and carbonate reservoir rocks. Wettability alteration in the case of sandstone reservoirs by LSWF is strongly influenced by the different type of clay particles present in the reservoir. The uneven distribution of clays throughout the sandstone reservoir rocks have also made it difficult to study the exact impact of each clay particle and that the amount of clay content has on wettability alteration. The major clay particles involved in wettability alteration mechanisms in sandstone reservoirs are montmorillonite, kaolinite and illite [5]. The clay particles

possess a net negative charge and they are able to adsorb polar components of crude oil (asphaltenes and resins) and thereby alter the rock surface to an oil-wet state. During the process of LSWF, cations are exchanged between the clay minerals and low salinity brines. The lower concentration of cations present in injected brine causes the equilibrium established between connate water and reservoir rocks to be disturbed, resulting in the cation exchange reactions. The cation exchange reactions result in desorption of polar components of oil from negatively charged clay surfaces and they are replaced by the cations from low salinity brine. The wettability of sandstone reservoir is found to be much more water-wet after the cation exchange process [6].

Thus, the process of cation exchange is another important factor that determines the extent of wettability alteration and thereby the amount of oil recovered from a sandstone reservoir. Different clay particles have different cation exchange capacity (CEC) and, based on their value of CEC, they are able to exert influence on the cation exchange process. Temperature and pressure have also been observed to affect the extent of wettability alteration by LSWF.

In addition to above mentioned mechanisms of enhanced oil recovery by LSWF, the results of experiments performed by Tang and Morrow [7] suggested that LSWF is able to cause the detachment of mobile clay particles (fines) such as kaolinite from the pore walls and thereby also carry the oil attached to pore walls through the clay particles, hence increasing the oil recovery, while high salinity brines do not disturb the clay particles attached to the pore walls.

The laboratory experiment conducted in this research project is designed to provide results that verify the main objective mentioned above and to also provide inferences to the following sub-objectives:

- To evaluate the role of content (concentration) of individual clay particles such as montmorillonite and kaolinite in modifying the wettability of sandstone rocks.
- To assess the role of reservoir temperature in the wettability alteration process of sandstone rocks.
- To determine the ideal low salinity concentration of injected brine required to obtain optimum oil recovery in sandstone reservoirs, by measuring contact angles of Sandstone samples before and after injection of three low salinity brines.
- To evaluate the pattern of variation in the values of contact angles measured for each sandstone sample (having different clay content) that are injected with three brines of varying low salinity concentrations.

## 2. Material and Methods

### 2.1. Materials

#### 2.1.1. Solids

The solid materials used in the experiment include mica powder, two clays and a salt. Mica powder is chosen to be the representative sandstone rock sample in order to diminish rock heterogeneity and hysteresis on readings taken from Drop Shape Analyser, DSA 100, machine (KRÜSS, Hamburg, Germany) that indicate the contact angle of each sample. Mica powder was supplied by CEMCRAFT (Bromsgrove, UK). The two clays used in this study are kaolinite and montmorillonite. Both kaolinite and montmorillonite were supplied by Sigma Aldrich (Irvine, UK). The salt used in this experiment to produce low salinity brine was sodium chloride (NaCl). Sodium chloride was provided by Fisher Chemicals (Loughborough, UK) with purity of 99.5%.

#### Kaolinite

Kaolinite is 1:1 type clay and contains a tetrahedral sheet of silicon ( $\text{SiO}_4$ ) and an octahedral sheet of aluminium in one layer and the layers are placed sequentially one above the other [8]. The layers are bound to each other by hydrogen bonds that exist between oxygen atoms in tetrahedral sheets and hydroxyl groups in octahedral sheets [9]. Kaolinite is not chemically inert and does possess a small

net negative charge due to broken bonds at the edges of the kaolinite particles. These small negative charges are stabilised by minor cationic substitutions. The least reactive clay particle in sandstone reservoirs is kaolinite, and it is also classified as a non-swelling type of clay particle. Due to less surface area and less substitutions occurring, they have low CEC and hence low reactivity [10].

### Montmorillonite

Montmorillonite clay belongs to the Smectite category of clays and is a 2:1 type of clay. Two sheets of tetrahedral silicon atoms surround a central octahedral sheet of aluminium atom [8]. Polar molecules such as water can occupy the space in between the tetrahedral and octahedral sheets in between the layers, leading to expansion of the overall clay particle [8]. Montmorillonite clay is also referred to as a swelling clay. Substitution of  $\text{Si}^{4+}$  by  $\text{Al}^{3+}$  in tetrahedral layers and  $\text{Al}^{3+}$  by  $\text{Mg}^{2+}$  in the octahedral layer results in a strong net negative charge for montmorillonite clays. The negative charge so developed is balanced by adsorbing exchangeable cations such as  $\text{Ca}^{2+}$  or  $\text{Na}^{2+}$  from the connate water [9]. Montmorillonite also possesses the highest CEC among all the clay particles found in sandstone reservoirs and montmorillonite is classified as a swelling type of clay.

#### 2.1.2. Liquid

Crude oil and distilled water are the two liquids that were used to study the wettability alteration process in mica powder mixed with clays (representing a sandstone sample). Crude oil was supplied by Statoil (Stavanger, Norway) and the source of the crude oil is the North Sea. The crude oil has an API gravity of 37.50. The distilled water was used in the preparation of low salinity brines. In this study, three types of low salinity brines with salinity content of 500 ppm, 1000 ppm and 2000 ppm were used to study the wettability alteration characteristic in sandstone samples.

## 2.2. Methods

### 2.2.1. Clay Content and Sandstone Reservoir Rocks

In this experiment, mica powder was employed to represent sandstone reservoir rock as it is one of the major components found in sedimentary rocks. It was used as the mean solid concentration and mixed with different amounts of kaolinite and montmorillonite separately. The preparation of samples by mixing mica powder and a single clay component (kaolinite/montmorillonite) was carried out to analyze the effect of that individual clay component on wettability alteration of sandstone rocks. Firstly, a total of eight samples were prepared by varying different concentrations of mica powder and the two clays. The first four samples contained either 15% or 30% clay content of one particular type of clay (kaolinite/montmorillonite) and was prepared at 25 °C. The remaining four also contained 15% or 30% clay content of one particular type of clay (kaolinite/montmorillonite) and was mixed at 50 °C. Then, the mixture of mica powder (sandstone samples) and clays that are naturally water-wet need to be altered to the oil-wet state by using crude oil. Finally, the contact angles of all eight samples were measured. The composition of all eight samples with varying clay content (15% or 30%) and type of clay are illustrated below in the table below (Table 1).

**Table 1.** Composition of eight samples with different concentrations of mica, kaolinite and montmorillonite.

Sample No.	S1	S2	S3	S4	S5	S6	S7	S8
Temperature (°C)	25	25	25	25	50	50	50	50
Clay Content (%)	15	30	15	30	15	30	15	30
Components								
	Mica (g)	42.5	35	42.5	35	42.5	35	42.5
	Kaolinite (g)	7.5	15	0	0	7.5	15	0
	Montmorillonite (g)	0	0	7.5	15	0	0	7.5

### 2.2.2. Oil-Wet Sample Restoration

Following procedure was applied to prepare oil-wet samples:

- (1) The powdered mix were measured in a digital weighing balance and the amount of powdered mix after the process of filtration recovered was noted as shown in the table below (Table 2).
- (2) The Crude oil and powdered mix of each sample was mixed in a fixed ratio. The ratio taken is for every 10 g of powdered mix, and 40 mL of crude oil was added. Based on this ratio, the amount of crude oil required for each sample is illustrated in the table below (Table 3).
- (3) The samples and required crude oil were mixed in 400 mL beakers and were stirred using a magnetic hot plate stirrer and magnetic beads. The stirring and heating (at 25 °C/50 °C) of each sample were as per specifications of the table above. The stirring was done to ensure proper mixing in order to obtain oil-wet samples. The mixture was allowed to stir at a medium rotation speed (45 rpm) and the stirring process was continued overnight for a duration of 24 h.
- (4) Two days of decantation time was required for all samples after the agitation process.
- (5) The mixture of crude oil and samples were then filtered using a vacuum pump, 1.5 L Erlenmeyer, a funnel and filter paper. All equipment mentioned here was cleaned and sterilized. The process of filtration for each sample took about two hours each. The solid phase of mixture was collected and then placed onto a watch glass.
- (6) The solid mixtures of samples obtained from the vacuum filtration process were then allowed to dry off in an oven preheated to 100 °C. The samples were allowed to dry in the oven for a time period of 24 h.
- (7) The dried samples were then removed from the oven and placed outside to cool off for 30 min. Finally, the solid mixtures of each sample were then crushed in a mortar to obtain powdered mix in order to measure contact angle of each sample.

**Table 2.** The powdered mix of each sample recovered after filtration process.

Sample No.	S1	S2	S3	S4	S5	S6	S7	S8
Temperature (°C)	25	25	25	25	50	50	50	50
Clay Content (%)	15	30	15	30	15	30	15	30
Powdered Mix Recovered (g)	38.5	36.3	37.7	25	47.2	47.2	41.3	27

**Table 3.** Quantity of crude oil required for each sample.

Sample No.	S1	S2	S3	S4	S5	S6	S7	S8
Temperature (°C)	25	25	25	25	50	50	50	50
Clay Content (%)	15	30	15	30	15	30	15	30
Powdered Mix (g)	38.5	36.3	37.7	25	47.2	47.2	41.3	27
Crude oil Required (mL)	160	145	150	100	189	189	165	108

### 2.2.3. Wettability Study of Oil Wetted Samples with Low Salinity Brines

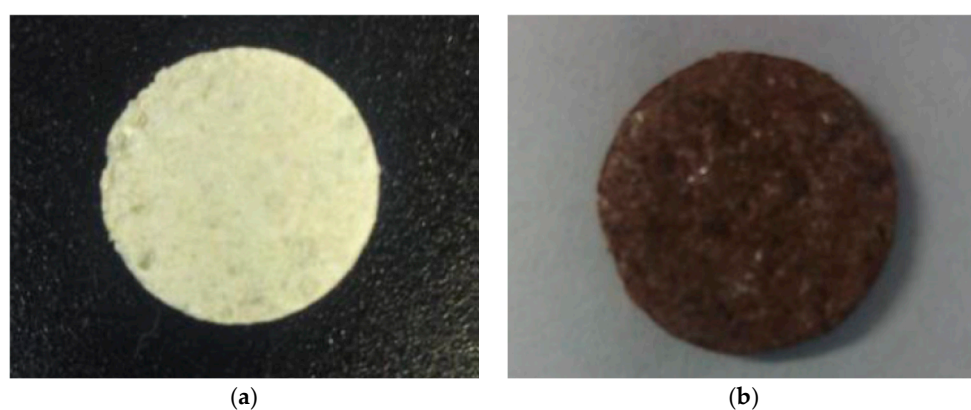
- (1) The prepared oil-wet samples from the above procedure were then used to study effect and extent of LSWF in the presence of clay on their wetting state.
- (2) The oil-wet powder mix was weighed in a digital weighing machine having a capacity of 250 g and minimum value of 0.01 g. The oil-wet powder mix of each sample was then equally divided into three different sets that are to be injected with the three brines. The mass of each sample to be mixed with each brine was determined using a fixed ratio. The ratio was fixed, as for every 4 g of oil-wet sample, 20 mL of brine was required. Based on this ratio, a total of 24 samples were prepared by mixing the required amount of the original eight oil-wet samples (S1–S8) and the

three brines (B1–B3) individually. For example, 20 g of oil-wet sample S1 was mixed with 100 mL of brine B1. The amount of brine required per sample was calculated and the required amount of oil-wet sample/brine is shown in the tables below (Table 4).

**Table 4.** Quantity of sample and each brine required for the tests.

Sample No.	S1	S2	S3	S4	S5	S6	S7	S8
Temperature (°C)	25	25	25	25	50	50	50	50
Clay Content (%)	15	30	15	30	15	30	15	30
Oil-wet Powder Available/Sample (g)	20	20	20	15	25	25	22	15
B1 Required (mL)	100	100	100	75	125	125	125	75
B2 Required (mL)	100	100	100	75	125	125	125	75
B3 Required (mL)	100	100	100	75	125	125	125	75

- (3) The beakers containing the required amount of oil-wet sample and brine were then placed on magnetic hot plate stirrers. A magnetic bead was placed in each beaker for stirring the mixture and stirring was done at medium speed of rotation (45 rpm). Heat was also provided at 25 °C for brines mixed with samples S1–S4, and brines mixed with samples S5–S8 were heated at 50 °C.
- (4) After stirring the samples for a time period of 2 h, they were removed from the hot plate stirrers and kept for two days for the process of decantation to occur.
- (5) After two days, the samples were filtered using a simple conical flask, funnel and filter paper. The flasks and funnels were thoroughly cleansed before being used for the filtration process. The filtration process takes about 1 h for each sample. The solid phase of each sample was then collected onto a watch glass.
- (6) The oven was preheated to 100 °C and the samples placed on the watch glass were kept in the oven and allowed to dry for 24 h.
- (7) The samples were then removed from the oven and placed outside to cool for a time period of 30 min. The solid mix of each sample was then crushed in a mortar to obtain the powder mix.
- (8) The powdered mixes of all 24 samples obtained were then used to make pellets and measure their contact angle as per the same procedure mentioned in the earlier section. Two examples of pellet discs made for contact angle measurement are presented in Figure 1.

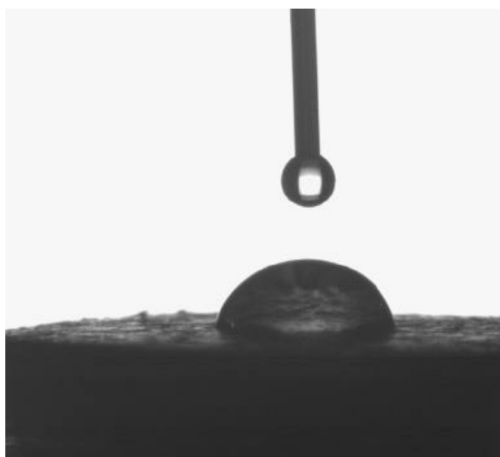


**Figure 1.** Example of discs made from powder mix for contract angle measure. (a) mica mixed with kaolinite; and (b) mica mixed with kaolinite and aged in crude oil.

#### 2.2.4. Contact Angle Measurements

The measurement of contact angle of each sample of oil-wet mica powder and clay mix was carried out using a Kruss drop shape analyser (DSA 100, KRÜSS, Hamburg, Germany). The contact angle

of sample was measured on a system with three interfaces, i.e., air, water and surface of powdered sample. The DSA 100 machine is regarded as the finest machine available to measure contact angle of samples with maximum accuracy. The final reported contact angle is the average of three contact angle measurements on the same surface. All contact angles were measured after 10 min of the releasing of droplets on the surface. Figure 2 below illustrates the type of images recorded by the DSA 100 machine for each sample.



**Figure 2.** Example of images of water droplets recorded by Drop Shape Analyser, DSA 100, on the rock sample.

### 3. Results and Discussion

#### 3.1. Role of Clay Content in Modifying Wettability of Sandstone Rocks to Oil-Wet

The influence of clay particles montmorillonite and kaolinite in modifying the wettability of naturally water-wet mica powder (representing sandstone reservoir) to the oil-wet state was analyzed by measuring the contact angles of the eight samples. Each sample's contact angles were measured after they had been altered with crude oil. Table 5 provides the composition of each sample and the result of the average value of contact angle obtained.

**Table 5.** Contact angle values of oil-wet sandstone samples.

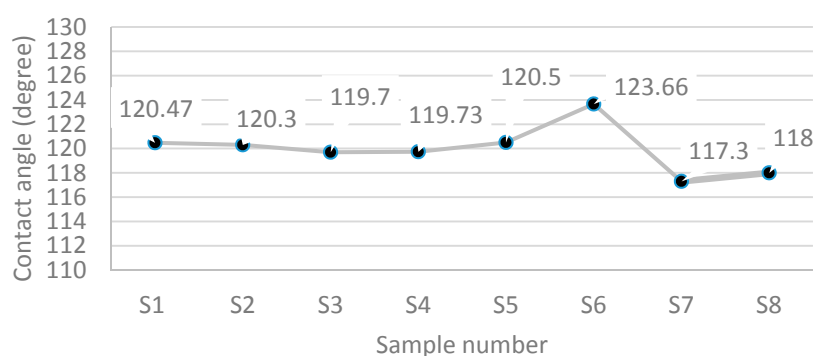
Sample No.	S1	S2	S3	S4	S5	S6	S7	S8
Temperature (°C)	25	25	25	25	50	50	50	50
Clay Type	Kaolinite		Montmorillonite		Kaolinite		Montmorillonite	
Clay Content (%)	15	30	15	30	15	30	15	30
Contact Angle Measured—Average Value	120.4°	120.3°	119.7°	119.7°	120.5°	123.6°	117.3°	111.0°

The measured contact angle values of sandstone samples containing both types of clays, i.e., kaolinite (S1, S2, S5 and S6) and montmorillonite (S3, S4, S7 and S8), indicate the successful alteration of wettability of sandstone from a naturally water-wet state to a more oil-wet state. The interaction of crude oil and clay particle montmorillonite in samples S3, S4, S7 and S8 has led to polar components of crude oil such as asphaltenes and resins being adsorbed onto the clay surface and modifying wettability to an oil-wet state [11]. In the case of sandstone samples S1, S2, S5 and S6 containing kaolinite clay particle, interaction between crude oil and kaolinite again led to the adsorption of polar components of crude oil and thereby alteration of wettability to an oil-wet state. Kaolinite is rich in hydroxyl



groups on its surface and thus is susceptible to adsorption of asphaltenes [12]. The adsorption of heavy components of crude oil such as asphaltenes and resins tend to impart a more hydrophobic nature to the clay particles [13].

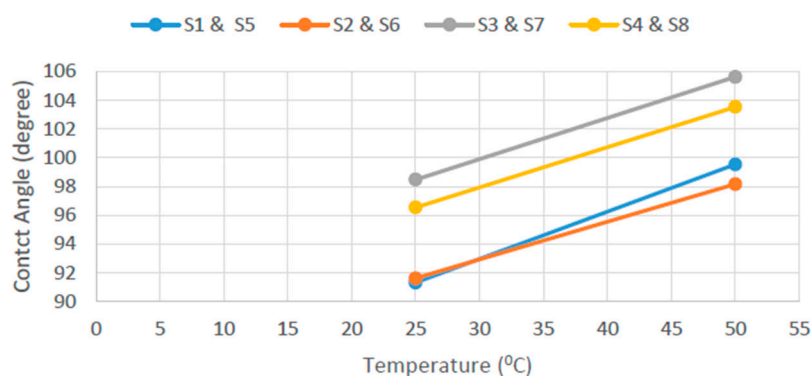
It is also observed that all four oil-wet samples having kaolinite clay content (S1, S2, S5 and S6) have higher contact angle values than their corresponding four oil-wet samples containing montmorillonite clay content (S3, S4, S7 and S8). Higher contact angle values of samples with kaolinite clays are due to the fact that kaolinite clay is hydrophobic in nature and shows more attraction towards asphaltenes and resins than montmorillonite clays [14]. Montmorillonite, on the other hand, is hydrophilic in nature and hence samples with montmorillonite have lower contact angle values when compared to samples with kaolinite content as shown in Figure 3 below.



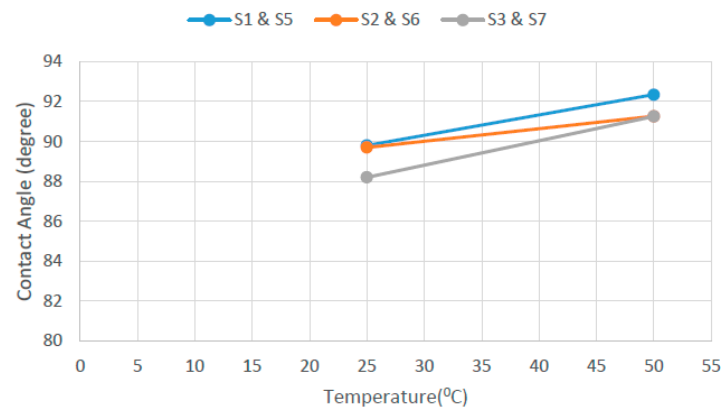
**Figure 3.** Contact angle values of samples S1–S8.

### 3.2. Effect of Temperature

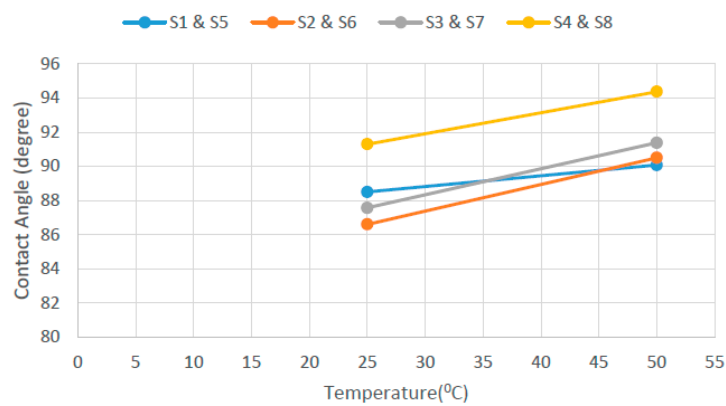
As shown in Figure 3 above, the contact angle values obtained do not show a significant variation in the case of samples S1, S2, S3 and S4 prepared at 25 °C. Samples S1 and S2 contain 15% and 30% of kaolinite clay. S3 and S4 contain 15% and 30% of montmorillonite clay, respectively. However, in the case of samples S5, S6, S7 and S8 (blended at 50 °C), we observe variation in contact angles measured. Samples S1 and S5 contain 15% kaolinite clay, whereas samples S2 and S6 contain 30% kaolinite clay. Though all four samples contain kaolinite clay, the increased temperature (50 °C) in the case of S5 and S6 has resulted in increased value of contact angles measured when compared to samples S1 and S2. The effect of increased temperature resulted in decreased contact angle values in the case of samples S5 and S6 (blended at 50 °C) that contain 15% and 30% montmorillonite clay when compared to samples S3 and S4 (blended at 25 °C). Figures 4–6 illustrate the increase in contact angle measured.



**Figure 4.** Contact angle variation in eight samples with increase in temperature for brine B1.



**Figure 5.** Contact angle variation in eight samples with increase in temperature for brine B2.



**Figure 6.** Contact angle variation in eight samples with increase in temperature for brine B2.

In the case of brine B1, the average difference in contact angle measured was about seven degrees for samples containing same type of clay and similar content (i.e., S1 and S5, S2 and S6, S3 and S7 and S4 and S8), as shown in Figure 4 above. In the case of brine B2, the average difference in contact angle measured was about three degrees again for samples with the same type of clay and similar clay content, whereas in the case of brine B3, the average difference in contact angle measured was about three to four degrees as shown in Figures 5 and 6 above.

The higher contact angle values indicate that the wettability alteration process at higher temperatures results in weakly water-wet state. Lower reservoir temperatures, on the other hand, seem to encourage a higher degree of wettability alteration from an oil-wet state to a strongly water-wet state [15]. The increase in temperature and decrease in degree of wettability alteration, observed in all cases except for the case of sample S8 injected with brine B2, demonstrates that the results of this experiment confirm the correlation between reservoir temperature and modification of wettability state.

### 3.3. Role of Low Salinity Brines in Wettability Alteration of Oil-Wet Samples

The eight oil-wet samples (S1–S8) were separately injected with three low salinity brines—B1, B2 and B3, respectively. Three contact angle measurements were taken and their average value was calculated and selected as the final contact angle value of each sample. The primary aim of this part of the experiment was to analyze the extent of wettability alteration to the water-wet state by measuring reduction in contact angle values for each sample when they were subjected to treatment with three brines having different salinity concentrations. The results also help to identify the ideal salinity range for LSWF operations. Table 6 provides the contact angle values obtained after low salinity water treatment for each brine. Treating oil-wet sandstone samples with the low salinity brines resulted



in reduction of contact angle values in the case of each sample (S1–S8). The values of contact angles obtained as shown in Table 6 lies in the intermediate-wet state [16]. The results indicate that the low salinity brines successfully altered the wettability of sandstone from an oil-wet state to a more water-wet state.

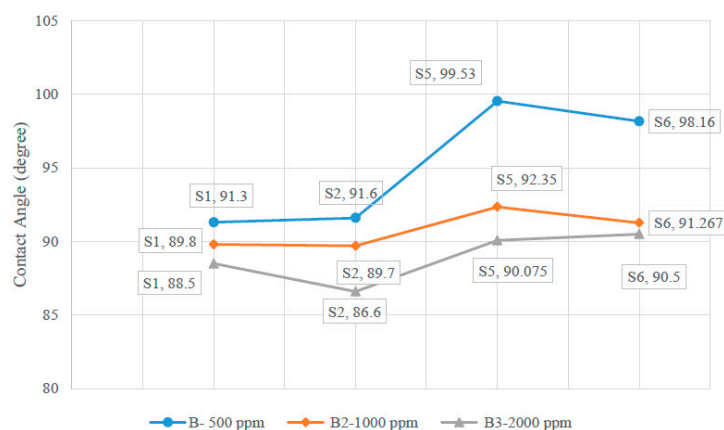
**Table 6.** Contact angle values of eight samples before and after injection with brines B1, B2 and B3.

Sample No.	Temperature (°C)	Contact Angle Average Value (°)			
		Oil-Wet State	Brine B1 500 ppm	Brine B2 1000 ppm	Brine B3 2000 2000 ppm
S1	25	120.467	91.3	89.8	88.5
S2	25	120.30	91.6	89.7	86.6
S3	25	119.70	98.47	88.2	87.567
S4	25	119.73	96.53	94.67	91.3
S5	50	120.50	99.53	92.35	90.075
S6	50	123.66	98.16	91.267	90.5
S7	50	117.30	105.63	91.26	91.38
S8	50	111.0	103.54	88.45	94.375

### 3.4. Effect of Type and Content of Clay Particle in Wettability Alteration

The results clearly indicate that samples containing clay particle kaolinite (S1, S2, S5 and S6) produced lower values of contact angle after treatment with brines B1, B2 and B3 as compared to the values obtained by samples containing montmorillonite clay (S3, S4, S7 and S8). The increase in clay content from 15% to 30% did not produce much variation in the values of contact angle measured for samples containing kaolinite clay (S1, S2, S5 and S6) in case of the three brines. However, in the case of samples containing montmorillonite clay (S3, S4, S7 and S8), the increase in concentration of clay content from 15% to 30% resulted in variation of contact angle values measured in the case of each brine as illustrated in Table 5. There is a difference of three degrees on average between measured contact angle values of samples S3 and S4 (mixed at 25 °C) containing 15% and 30% of Montmorillonite clay in the case of the three brines B1, B2 and B3. Similarly, there is a difference of three degrees on average between measured contact angle values of samples S7 and S8 (mixed at 50 °C) containing 15% and 30% of montmorillonite clay in the case of the three brines B1, B2 and B3.

Figures 7 and 8 further illustrate the variation in contact angle observed for samples with varying concentration of clay content when treated with different brines. The values of contact angle measured for all three brines lie in the intermediate-wet range, i.e., 75°–115° [17]. The highest recovery factors have been observed for reservoirs having intermediate-wet state of wettability [18]. The wettability modification of samples to the intermediate-wet state by low salinity brines in the experiment conducted here indicates that low salinity brines are able to yield the highest recovery factors when applied as a secondary recovery method.



**Figure 7.** Variation in contact angle of samples containing kaolinite clay with each brine.

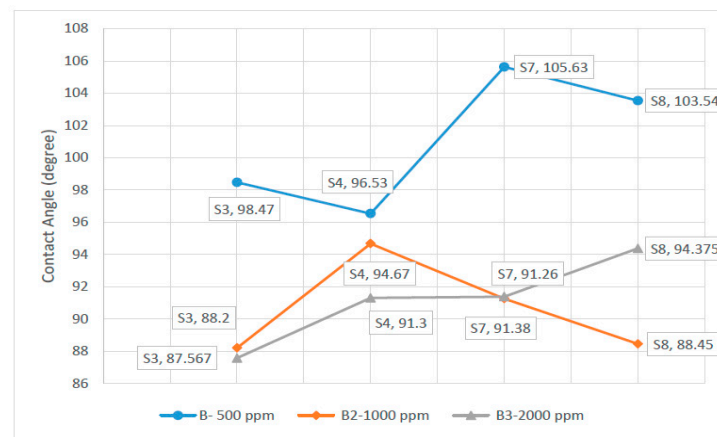


Figure 8. Variation in contact angle of samples containing montmorillonite clay with each brine.

### 3.5. Effect of Salinity Concentration of Brine in Wettability Alteration

All three brines (B1, B2 and B3) managed to alter the wettability of each sample (S1–S8) from an oil-wet state to a more water-wet state as shown by the measured contact angle values in Table 6. The lowest value of contact angle of six samples (S1, S2, S3, S4, S5 and S6) was measured in the case of brine B3, having a salinity concentration of 2000 ppm. The lowest value of contact angle measured for samples S7 and S8 was for the case of brine B2, having a salinity concentration of 1000 ppm. The highest values of contact angle measured for each sample was in the case of brine B1. Figure 9 below illustrates the variation in contact angle of each sample with the three brines.

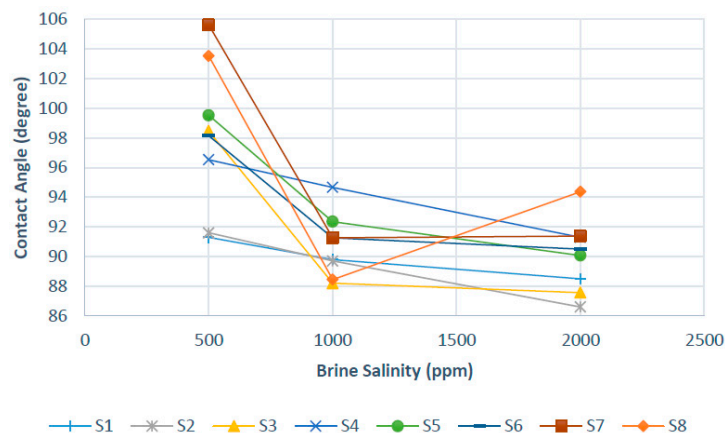


Figure 9. Contact angle value of each sample for brines B1, B2 and B3.

The ideal salinity range of brines to be injected for low salinity water injection operations was identified in the literature review section as lying in the range of 1000 and 2000 ppm. This ideal salinity range was first mentioned as a pre-requisite condition in order to observe wettability alteration caused by the low salinity brines by the same researchers who proposed a chemical mechanism to explain low salinity effect [19]. The results of the experiment conducted here clearly demonstrate that the lowest contact angle values and hence the highest degree of wettability alteration were observed when salinity concentration of injected brine was in the range of 1000–2000 ppm (B2 and B3). Thus, the results obtained in this experiment provided further evidence that the ideal salinity range for improved oil recovery by LSWF is between 1000 and 2000 ppm.

#### 4. Conclusions

- (1) The results of the laboratory experiment conducted in this work provides further insight and confirmation that the influence that clay particles, temperature and salinity concentration has on wettability of sandstone rocks when treated with low salinity brines. The experiment involved the use of samples containing mica powder, different type of clay particles (kaolinite/montmorillonite) and varying clay content (15%/30%) that represented sandstone reservoir rock samples. These samples were modified with crude oil from the North Sea and then treated with three brines of sodium chloride having varying low salinity concentration (500, 1000 and 2000 ppm). The main conclusions that can be drawn from the results of the experiment conducted are listed below.
- (2) The modification of mica powder and clay mix with crude oil proved that the type of clay and clay content do play a significant role. Samples containing kaolinite clay produced higher contact angle values when compared to samples with montmorillonite clay.
- (3) The contact angle values measured indicate wettability alteration of oil-wet samples to intermediate-wet state as the contact angle values lie in the range of 75°–115°. Thus, low salinity brines when applied as injection brine are able to modify wettability to the intermediate-wet range and thereby produce the highest oil recovery rates, as reservoirs with a wettability state of intermediate-wet have the highest recovery factors.
- (4) The salinity range established for observing the highest degree of wettability alteration from the oil-wet state to a more water-wet state in the experiment conducted here is 1000–2000 ppm. Thus, the ideal salinity concentration for low salinity brine lies in the range 1000–2000 ppm.
- (5) The results of the experiment further verify the established correlation between reservoir temperature and the extent of wettability alteration, i.e., the higher the reservoir temperature, the lower the degree of wettability alteration from an oil-wet state to a more water-wet state.

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#### References

1. Jerauld, G.R.; Webb, K.J.; Lin, C.Y.; Secombe, J.C. Modeling low-salinity waterflooding. *SPE Reserv. Eval. Eng.* **2008**, *11*, 1000–1012. [[CrossRef](#)]
2. Mahani, H.; Keya, A.L.; Berg, S.; Bartels, W.B.; Nasralla, R.; Rossen, W.R. Insights into the mechanism of wettability alteration by low-salinity-flooding (LSF) in carbonates. *Energy Fuels* **2015**, *29*, 1352–1367. [[CrossRef](#)]
3. Nasralla, R.A.; Sergienko, E.; Masalmeh, S.K.; Van der Linde, H.A.; Brussee, N.J.; Mahani, H.; Suijkerbuijk, B.; Alqarshubi, I. Demonstrating the potential of low-salinity waterflood to improve oil recovery in carbonate reservoirs by qualitative coreflood. In Proceedings of the SPE Abu Dhabi International Petroleum Exhibition and Conference, Abu Dhabi, UAE, 10–13 November 2014.
4. Law, S.; McDonald, A.; Fellows, S.; Reed, J.; Sutcliffe, P.G. Influence of clay content and type on oil recovery under low salinity waterflooding in North Sea reservoirs. In Proceedings of the SPE Offshore Europe Conference and Exhibition, Aberdeen, UK, 8–11 September 2015.
5. Grim, R.E. *Clay Mineralogy*, 2nd ed.; McGraw-Hill: New York, NY, USA, 1953.
6. Berg, S.; Cense, A.W.; Jansen, E.; Bakker, K. Direct experimental evidence of wettability modification by low salinity. *Petrophysics* **2010**, *51*, 314–322.
7. Tang, G.; Morrow, N. Salinity, temperature, oil composition and oil recovery by waterflooding. *SPE Reserv. Eng.* **1997**, *12*, 269–276. [[CrossRef](#)]

8. Shehata, A.M.; Nasr-El-Din, H.A. Zeta potential measurements: Impact of salinity on sandstone minerals. In Proceedings of the SPE International Symposium on Oilfield Chemistry, The Woodlands, TX, USA, 13–15 April 2015.
9. White, G.N.; Dixon, J.B. Kaolin-Serpentine Minerals. In *Soil Mineralogy with Environmental Applications*; Dixon, J.B., Schulze, D.G., Eds.; Soil Science Society of America: Fitchburg, WI, USA, 2002; pp. 389–414.
10. Bhattacharyya, K.G.; Gupta, S.S. Adsorption of a few heavy metals on natural and modified kaolinite and montmorillonite: A review. *Adv. Colloid Interface Sci.* **2008**, *140*, 114–131. [[CrossRef](#)] [[PubMed](#)]
11. Clementz, D.M. Interaction of petroleum heavy ends with montmorillonite. *Clays Clay Miner.* **1976**, *24*, 312–319. [[CrossRef](#)]
12. Bantignies, J.L.; Cartier Dit Moulin, C.; Dexpert, H. Wettability contrasts in kaolinite and illite clays: Characterization by infrared and X-ray absorption spectroscopies. *Clays Clay Miner.* **1997**, *45*, 184–193. [[CrossRef](#)]
13. Clementz, D.M. Clay stabilization in sandstones through adsorption of petroleum heavy ends. *J. Pet. Technol.* **1977**, *29*, 1061–1066. [[CrossRef](#)]
14. Borysenko, A.; Clennell, B.; Sedev, R.; Burgar, I.; Ralston, J.; Raven, M.; Dewhurst, D.; Liu, K. Experimental investigations of the wettability of clays and shales. *J. Geophys. Res. Solid Earth* **2009**, *114*. [[CrossRef](#)]
15. Nasralla, R.A.; Bataweel, M.A.; Nasr-El-Din, H.A. Investigation of wettability alteration by low salinity water. In Proceedings of the Offshore Europe Meeting, Aberdeen, UK, 6–8 September 2011.
16. Anderson, W.G. Wettability literature survey—Part 1: Rock/oil/brine interactions and the effects of core handling on wettability. *J. Pet. Technol.* **1986**, *10*, 1125–1144. [[CrossRef](#)]
17. Anderson, W.G. Wettability literature survey—Part 2: Wettability measurement. *J. Pet. Technol.* **1986**, *11*, 1246–1262. [[CrossRef](#)]
18. Morrow, N.R. Wettability and its effect on oil recovery. *J. Pet. Technol.* **1990**, *42*, 1476–1484. [[CrossRef](#)]
19. Austad, T.; Rezaeidoust, A.; Puntervold, T. Chemical mechanism of low salinity water flooding in sandstone reservoirs. In Proceedings of the SPE Improved Oil Recovery Symposium, Tulsa, OK, USA, 24–28 April 2010.



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