

Effect of Temperature on Wettability of Oil/Brine/Rock Systems

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Supporting Information

ABSTRACT: We report results from a systematic investigation of the effect of the temperature on the wettability of oil/brine/rock systems. An oil sample, produced from a sandstone reservoir, was tested on sandstone-like substrates (i.e., mica and quartz) in NaCl and MgCl₂ solutions with concentrations ranging from 0 to 3 M. Raising the temperature from 25 to 50 °C has no discernible effect on the contact angle, regardless of substrate type, brine type, or salt concentration. Another oil sample, obtained from a carbonate reservoir, was examined on carbonate-like substrates (i.e., calcite) in NaCl and MgCl₂ solutions over a concentration range of 0–1 M. The contact angles decrease as the temperature increases from 25 to 65 °C, and this temperature effect also strongly depends upon the brine type and salt concentration. A systematic examination of the ζ potential of rock/brine and oil/brine interfaces under different conditions and subsequent discussions indicate that contact angle and ζ potential may not be directly linked. These findings regarding the wettability of oil/brine/rock systems may improve the understanding of low-salinity waterflooding mechanisms by elucidating the combined effects of the temperature and other critical variables, including brine type, brine concentration, crude oil composition, and substrate type.

1. INTRODUCTION

Low-salinity waterflooding (LSW) has received much attention as a potential technique for improved oil recovery as a result of its high efficiency and low capital and operating costs.^{1–7} LSW is often used in secondary and tertiary modes to facilitate the release of trapped oil. Studies have shown that LSW can be effective in both carbonates and sandstones when conditions are favorable.¹

Various mechanisms have been proposed to explain the improved oil recovery by LSW, and among these mechanisms, wettability alteration is generally regarded as a major factor.^{1–7} As the name implies, the injection of low-salinity brine into the oil reservoir may alter the wettability of reservoir rocks toward a state more favorable for oil recovery. Numerous studies have been conducted in the past 2 decades to investigate the wettability of oil/brine/rock from different perspectives, and the findings are summarized in a recent review.¹

The temperature may significantly affect wettability.⁸ Petke et al. studied the change of the contact angle of organic liquid droplets in equilibrium with saturated vapor on polymeric solid surfaces when the temperature varied between 5 and 160 °C and found a maximum contact angle change of 100°. The effect of the temperature on the wettability of oil/brine/rock, however, has received limited prior attention. Nasralla et al. examined the static contact angles between dead crude oils and five synthetic brine solutions of different total dissolved solids (TDS) on mica minerals at 140, 212, and 250 °F. In their work, the static contact angles are defined as the equilibrium angles after the droplets left the needle tip and stayed on the substrate surface. Nasralla et al. found that the contact angle measured through the denser water phase increased by 5–30° when the temperature increased, indicating that the mica surface became

more oil-wet at higher temperatures.² Wang et al. reported the static contact angle of two types of crude oils and synthetic brine solutions on calcite and quartz slides in a temperature window of 50–200 °F. They observed a 10° decrease in contact angle for calcite when the temperature increased from 70 to 200 °F, suggesting calcite surfaces became more water-wet as the temperature increased.³ Similar trends were also reported by later studies.⁴ Wang et al. showed that the quartz surface became more oil-wet when the temperature increased.³ In another study, however, the quartz surface remained a nearly constant wettability state at temperatures up to 162 °C and then became a complete oil-wet nature when the temperature increased from 162 to 200 °C.⁵ Alotaibi et al. measured the contact angles of petroleum fluid droplets and two different sandstones in three brine solutions, and they found that the influences of the temperature on wettability strongly depended upon the brine and mineral compositions. For instance, they found that, as the temperature increased from 122 to 194 °F, the contact angles for Berea sandstone increased ~13°, whereas the contact angles for Scioto sandstone decreased ~7°. In most of these previous investigations, the TDS of brine remained constant and the type and concentration of ion species were varied. The ion types and concentrations have been shown to affect the wettability of oil/brine/rock in different manners, and the observed overall effects might be due to a combination of multiple factors.⁷ There has been no previous work addressing the effect of the temperature on oil/brine/rock wettability in a systematic manner. Additionally,

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most previous studies simply reported the trends with few fundamental explanations provided.

To fill these gaps in the literature, we have embarked on a systematic investigation on the effect of the temperature on wettability of oil/brine/rock systems. The combined effects of the temperature and other critical variables, including brine type, brine concentration, crude oil composition, and substrate type, are explored. The ζ potentials at rock/oil and brine/oil surfaces under different conditions are also measured to provide insights.

2. MATERIALS AND METHODS

2.1. Materials. Two petroleum fluid samples provided by Chevron were used in this work, and we refer them as oils 1 and 2. Oil 1 is from a sandstone reservoir, and oil 2 is from a carbonate reservoir. Oils 1 and 2 were heated in a jacketed glass beaker for 24 h at 55 and 70 °C, respectively, to evaporate the lighter components in the crude oils. The heating temperatures were selected as 5 °C higher than the reservoir temperature of the corresponding crude oil (i.e., 50 °C for oil 1 and 65 °C for oil 2). The purpose of this heating step is to prevent bubble formation at high temperatures that might jeopardize the accuracy of contact angle measurements. Table 1 summarizes the

Table 1. Properties of Heated Oils 1 and 2

	oil 1	oil 2
density (g/mL)	0.9524	0.888
asphaltene content (wt %)	3.3	2.1
resin content (wt %)	36.8	15.9
TAN (mg of KOH/g of oil)	0.21	0.18
TBN (mg of equiv KOH/g of oil)	5.60	1.14
viscosity at 25 °C (cP)	2190	125
viscosity at 50 °C (cP)	336	32
viscosity at 65 °C (cP)		18.4

properties of heated oils 1 and 2. The total acid number (TAN) and total base number (TBN) were measured on the basis of ASTM D664 and D2896 standards, respectively.

Three types of substrates were employed: muscovite mica, quartz, and calcite. Mica and quartz slides were purchased from Tedpella, and calcite slides were provided by Chevron. Sodium chloride (i.e., NaCl) and magnesium chloride (i.e., MgCl₂) brine solutions were employed to assess the effect of mono- and divalent ions on wettability.

We used quartz and calcite particles to prepare the rock/brine mixtures for ζ potential measurements. These particles were obtained from Sigma-Aldrich. The quartz particles have a particle size less than 74 μm , and the calcite particles have a particle size less than 30 μm .

All chemicals used in this work (i.e., NaCl, MgCl₂, *n*-decane, and toluene) were analytical grade and obtained commercially.

2.2. Contact Angle Measurements. **2.2.1. Room-Temperature Measurements.** The procedure to measure the contact angle at room temperatures is similar to that reported previously.⁷ Briefly, 500 mL of brine solution of a desired concentration was loaded into a square-shape glass beaker. The substrate slide, supported by a customized stainless-steel holder, was submerged in the brine solution for 1 h. This aging time is adequate for the substrate slide to pre-equilibrate with brine. Then, we placed oil droplets on the substrate slide by the inverted pendant drop method, which has been widely used to measure the contact angle of the oil/brine/rock system.^{2,3,5,7} We first placed the needle tip close to the bottom surface of the substrate slide and slowly dispensed 100 μL volume of oil. The oil droplet touched the bottom of the substrate slide once it reached a certain size, and it remained in contact with the slide when the oil was being dispensed. We then gently pulled the needle downward, leaving the oil droplet to stay on the bottom surface of the substrate slide. The system was covered by plastic wrap to minimize the supply of air to the fluids in the container. The droplets were allowed to equilibrate for 3 days

before measurements were taken.⁷ The droplet images were captured by a 12 \times ThorLabs camera, and they were then analyzed by an ImageJ program “DropSnake” to obtain the contact angle. Note that the contact angle measured according to our procedure is the same as the water-receding contact angle, which is the dynamic contact angle measured during the process, in which the volume of an oil droplet in contact with substrate is slowly increased until the contact line moves outward.¹⁰ Four droplets were placed on the slides at each run, and we report the results by their average and standard deviation. We consistently used a 100 μL droplet volume to eliminate the influence of the droplet size on the contact angle.⁷

2.2.2. High-Temperature Measurements. We employed an environmental chamber (p/n 100-07) and a temperature controller (p/n 100-50) from Rame-Hart Instrument Co. to measure the contact angle at elevated temperatures. The temperature of chamber walls is controlled by the integral electrical heaters. The experiments were conducted in a quartz cell located inside the chamber to avoid the corrosion of chamber walls by brine at elevated temperatures. The substrate was fixed in the brine by a customized holder made of polytetrafluoroethylene (PTFE) Teflon. In a typical experiment, we first prepared the desired brine solution and degassed it for 5 min. This degassing step was employed to avoid the formation of air bubbles during contact angle measurements at elevated temperatures. The degassed brine was loaded into the quartz cell. We then filled the gap between chamber walls and quartz cell walls with deionized (DI) water to ensure sufficient heat transfer. Next, the chamber was sealed, and the substrate was aged in brine at the set-point temperature for 1 h. After the time had elapsed, we removed the chamber cover and placed three droplets on the substrate bottom surface by the inverted pendant drop method. The droplets were allowed to equilibrate for 3 days before measurements were taken. We analyzed the captured image of each droplet using the ImageJ program and report the results by their average and standard deviation.

2.3. ζ Potential Measurements. A ζ potential analyzer and particle sizer (ZetaPALS, Brookhaven Instruments) was employed to measure the ζ potential of rock/brine and oil/brine interfaces. To measure the ζ potential of the oil/brine interface, we mixed brine and oil at a 10:1 (v/v) ratio and sonicated the mixture in a bath sonicator for 30 s. To measure the ζ potential of the rock/brine interface, we mixed 0.1 g of solid particles in a 5 mL brine solution. Afterward, the mixture was sonicated in a bath sonicator for 30 s. All sonicated samples were left 2 h before measurements were taken. We have examined the temporal variation of the ζ potential of rock/brine and oil/brine interfaces for 3 days and noticed that the ζ potential became stable after 2 h in both cases. These results are presented in the Supporting Information. These trends imply that, after 2 h, the equilibrium state has been reached in our systems, whereas some other authors have reported longer equilibration times in their particular systems.⁶

In a typical ζ potential measurement, we loaded 1.5 mL of pre-prepared sample into the cuvette and carefully inserted the electrode to avoid creation of bubbles. The instrument was set to automatically select the appropriate magnitude and frequency of the applied electrical field based on the conductance of the sample. A total of 10 runs of 30 cycles were performed for each sample, and the results were reported as the average value and standard deviation. We replicated each test at least 2 times to ensure good reproducibility.

3. RESULTS AND DISCUSSION

This section first provides the effect of the temperature on the contact angle of oil/brine/rock systems. We then assess the ζ potential of rock/brine and oil/brine interfaces under different conditions. We conclude with a discussion of the ζ potential results, and the insights that they provide for contact angle and wettability.

3.1. Effect of the Temperature on the Contact Angle of Oil/Brine/Substrate Systems. **3.1.1. Effect of the Temperature on the Contact Angles of Oil 1/Brine/Mica**

and Oil 1/Brine/Quartz Systems. Oil 1 was produced from a sandstone reservoir, and thus, it was tested on mica and quartz, the two abundant components of sandstones.⁷ Figures 1 and 2

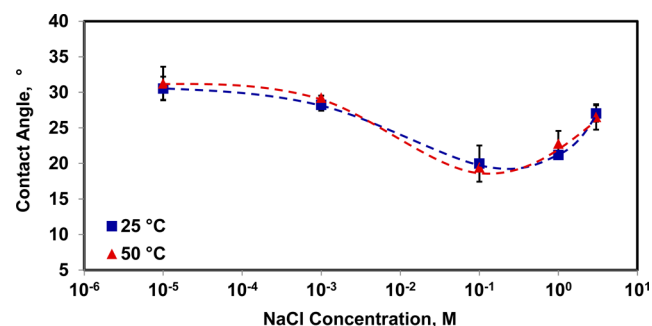


Figure 1. Effect of the temperature on the contact angles of the oil 1/NaCl/mica system. The error bars represent the standard deviation from different droplets.

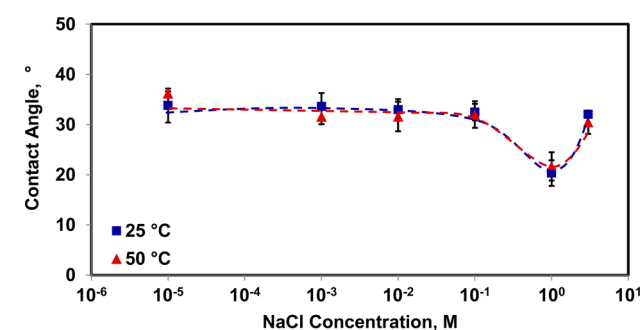


Figure 2. Effect of the temperature on the contact angles of the oil 1/NaCl/quartz system. The error bars represent the standard deviation from different droplets.

depict the contact angles of oil 1/NaCl/mica and oil 1/NaCl/quartz systems, respectively, under different conditions. The reservoir temperature for oil 1, 50 °C, was selected as the highest temperature examined. Dashed lines in each figure aid the identification of trends. Figure 1 shows that, as the NaCl concentration increases, the contact angles gradually decrease from $\sim 30^\circ$ in DI water to a minimum value of $\sim 20^\circ$ at 0.1 M and then climb back to $\sim 30^\circ$ when the NaCl concentration reaches 3 M. The trend of contact angle changing with the NaCl concentration is similar in the oil 1/NaCl/quartz system, as shown in Figure 2. The contact angles remain nearly constant below 0.1 M, decrease to a minimum value of $\sim 20^\circ$ at 1 M, and then increase back to $\sim 30^\circ$ when the NaCl concentration increases from 1 to 3 M. Bera et al. also measured the contact angles of an oil/NaCl/quartz system in a concentration range of 0–8 wt % NaCl. A minimum contact angle was observed at 6 wt % NaCl,¹¹ which is in a similar concentration range as the present study. In both oil 1/NaCl/mica and oil 1/NaCl/quartz systems, the contact angles are close at 25 and 50 °C, indicating that raising the temperature from 25 to 50 °C has no appreciable effect in these two systems.

Figures 3 and 4 display the contact angles of oil 1/MgCl₂/mica and oil 1/MgCl₂/quartz systems, respectively, under different conditions. The behaviors of mica and quartz are similar, but their responses to different salt concentrations are drastically different from those in NaCl. Figure 3 shows that, as the MgCl₂ concentration increases, the contact angles remain

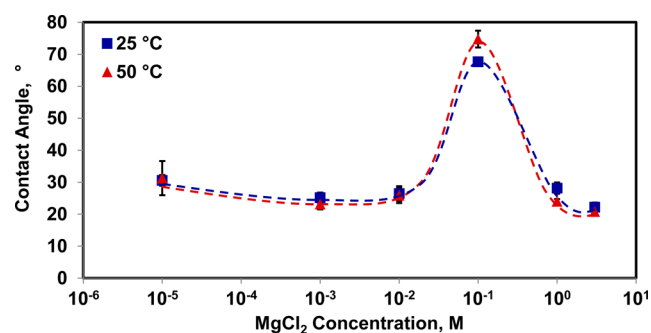


Figure 3. Effect of the temperature on the contact angles of the oil 1/MgCl₂/mica system. The error bars represent the standard deviation from different droplets.

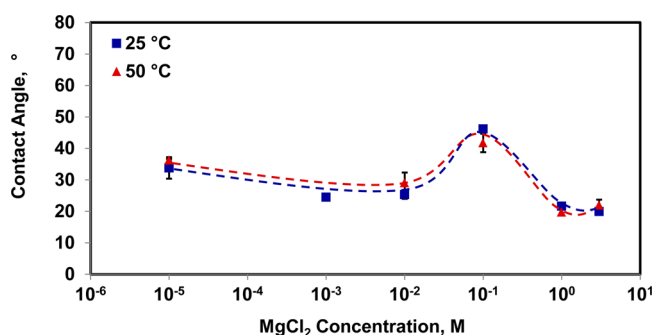


Figure 4. Effect of the temperature on the contact angles of the oil 1/MgCl₂/quartz system. The error bars represent the standard deviation from different droplets.

nearly constant below 0.01 M, sharply increase to a maximum value of $\sim 70^\circ$ at 0.1 M, then fall to $\sim 20^\circ$, and remain constant at higher concentrations. Quartz displays similar responses to the change of MgCl₂ concentrations as mica, but the maximum contact angle in oil 1/MgCl₂/mica is nearly 2 times higher than it is in oil 1/MgCl₂/quartz. Again, the contact angles measured at 25 °C are close to those at 50 °C, indicating that the change of the temperature in this range has little influence on the contact angles of oil 1/MgCl₂/mica and oil 1/MgCl₂/quartz systems.

3.1.2. Effect of the Temperature on the Contact Angles of the Oil 2/Brine/Calcite System. The above results reveal that raising the temperature has no appreciable effect on the contact angle of oil 1, a petroleum fluid produced from a sandstone reservoir, on sandstone-like minerals (i.e., mica and quartz) in both NaCl and MgCl₂ solutions over a concentration range of 0–3 M. Next, we present results of contact angles of oil 2, a petroleum fluid produced from a carbonate reservoir, on carbonate-like minerals (i.e., calcite) in both NaCl and MgCl₂ solutions over a concentration range of 0–1 M. Figures 5 and 6 depict the contact angles of oil 2/NaCl/calcite and oil 2/MgCl₂/calcite systems, respectively, under different conditions. As noted above, the reservoir temperature of oil 2 is 65 °C, and thus, this temperature was selected as the highest temperature examined for oil 2. Figure 5 shows that the contact angles increase monotonically with increasing NaCl concentrations, indicating a more oil-wetting status at a higher salinity regime. Initially, the contact angles increase slowly with increasing salt concentrations and then rapidly when the NaCl concentration exceeds 0.01 M. A comparison between the contact angles at room temperature and reservoir temperatures indicates that raising the temperature produces a more water-wet surface and

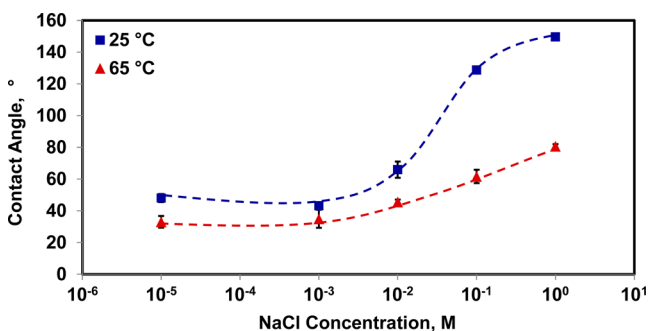


Figure 5. Effect of the temperature on the contact angles of the oil 2/NaCl/calcite system. The error bars represent the standard deviation from different droplets.

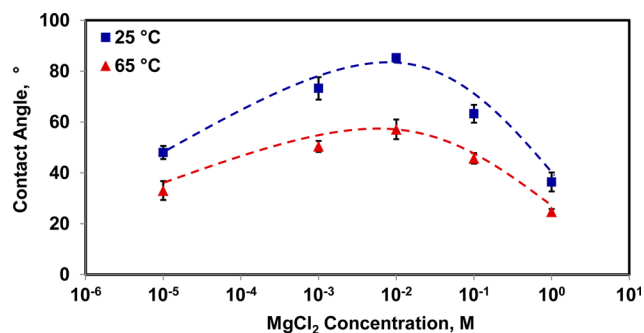


Figure 6. Effect of the temperature on the contact angles of the oil 2/MgCl₂/calcite system. The error bars represent the standard deviation from different droplets.

that this effect is more pronounced at higher NaCl concentrations. Figure 6 shows that, as the MgCl₂ concentration increases, the contact angles gradually increase to a maximum and then decrease. At higher temperatures, the contact angles become smaller, indicating a more water-wetting status. The difference between contact angles at room

temperature and reservoir temperatures remains nearly constant over the entire MgCl₂ concentrations investigated, suggesting that the temperature effect is independent of the MgCl₂ concentration.

3.2. ζ Potential of Rock/Brine and Oil/Brine Interfaces.

The contact angle reflects the complex microscopic interactions of the components of oil, rock, and brine. To further elucidate these interactions, we present our measurements of the ζ potential of rock/brine and oil/brine interfaces and then use these results to examine the variation of the contact angle under different conditions.

3.2.1. Effect of the Temperature on the ζ Potential of Rock/Brine Interfaces. Figure 7 displays the measurements of the ζ potential of rock/brine interfaces. The ζ potential of the quartz/NaCl interface initially becomes more negative with increasing NaCl concentrations at low NaCl concentrations and then less negative with increasing NaCl concentrations at higher NaCl concentrations. In contrast, the ζ potential of the quartz/MgCl₂ interface becomes increasingly less negative with increasing MgCl₂ concentrations. Silica, the primary component of quartz, undergoes extensive hydroxylation when it is in contact with water, and thus, the quartz surface carries negative charges.¹² When a salt, such as NaCl or MgCl₂, is added to the aqueous phase, the positive ions Na⁺ or Mg²⁺ may adsorb to the surface. In a recent paper, we demonstrate the adsorption of Na⁺ ions on the mica surface by molecular dynamics simulations.¹³ Despite the fact that mica and quartz have different chemical compositions, our results for mica indicate that Na⁺ and Mg²⁺ ions may also adsorb to the quartz surface. As a result, the ζ potential of quartz/NaCl and quartz/MgCl₂ interfaces become less negative as the salt concentration increases and even positive at a high-salinity regime. Figure 7 also shows that the rate of the change of ζ potential with increasing salt concentrations depends upon ion charge density. The divalent Mg²⁺ ions raise the ζ potential at a faster rate than Na⁺ ions, and the ζ potential of the quartz/MgCl₂ interface reaches an isoelectric point at lower salt concentrations.

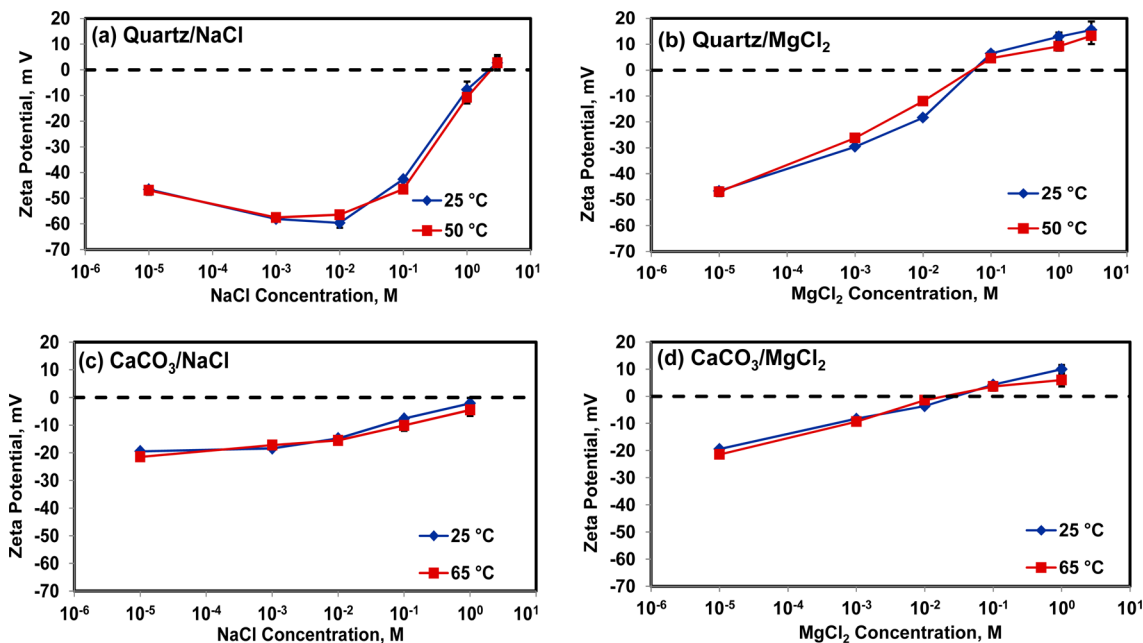


Figure 7. Effect of the temperature on the ζ potential of rock/brine interfaces: (a) quartz/NaCl, (b) quartz/MgCl₂, (c) CaCO₃/NaCl, and (d) CaCO₃/MgCl₂.

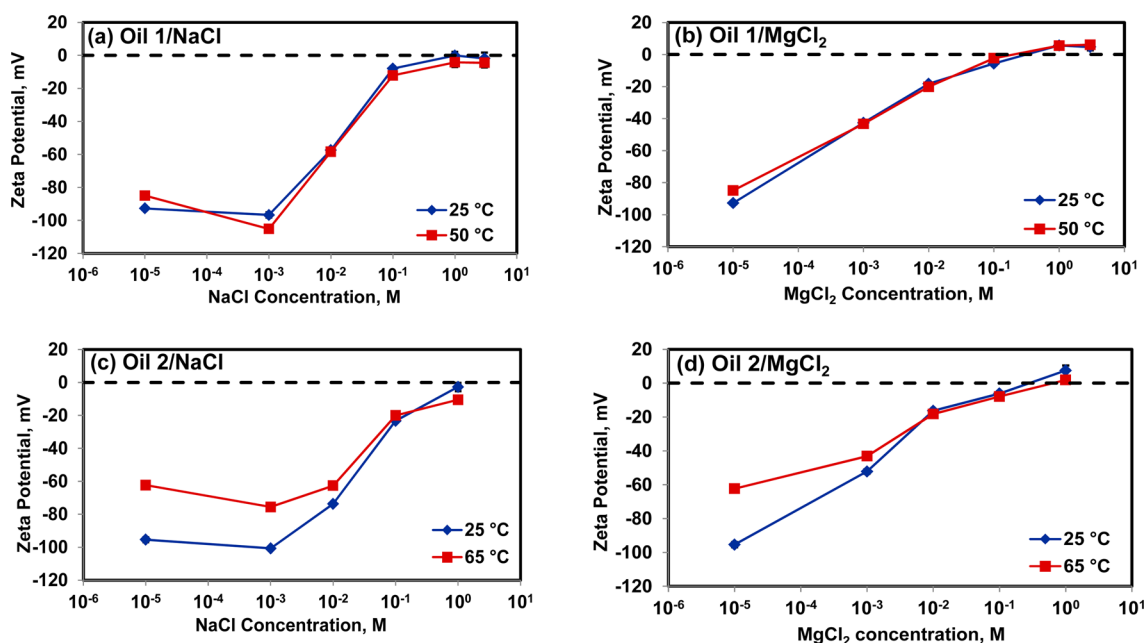


Figure 8. Effect of the temperature on the ζ potential of oil/brine interfaces: (a) oil 1/NaCl, (b) oil 1/MgCl₂, (c) oil 2/NaCl, and (d) oil 2/MgCl₂.

The trends of the ζ potential changing with brine concentrations in calcite/brine systems are similar to those in the quartz/brine systems. In the absence of salt, the ζ potential of the calcite/water interface is about -21 mV, much less negative than the ζ potential of the quartz/water interface. As a result of the very low solubility of calcium carbonate (the major component of calcite) in water, formation of CO_3^{2-} ions, hydrolysis, and other complicated reactions, the surface of calcite may undergo hydroxylation to a lesser extent than the quartz surfaces. As the salt concentration increases, more Na^+ or Mg^{2+} ions adsorb to the calcite surface, raising the ζ potential, as displayed in panels c and d of Figure 7. The temperature has little effect on the ζ potential of rock/brine surfaces under all of the conditions examined, probably because the temperature range investigated has no appreciable effect on ionization.

3.2.2. Effect of the Temperature on the ζ Potential of Oil/Brine Interfaces. Unlike the rock/brine interface, where the interactions between surface components and brine ions have been extensively investigated, the molecular interactions at the oil/brine interfaces have received less prior attention. A high negative ζ potential of nonpolar oils dispersed in brine has been reported,^{14,15} although there has been much debate in the literature on the source of these charges.^{16,17} Marinova et al. reported the average ζ potential of xylene droplets dispersed in aqueous solution to be around -80 mV at a low NaCl concentration of 10^{-5} M, and the ζ potential changed to -30 mV when the NaCl concentration reached 10^{-2} M. In the same study, the ζ potential of dodecane and hexadecane droplets dispersed in NaCl solution was also measured and their values were found to be around -60 mV at pH 6 and 10^{-3} M NaCl.¹⁴ Franks et al. reported the ζ potential of hexadecane-in-brine emulsions in the absence of surfactant to be in the range of -70 and -25 mV; the values depend upon the salt type, salt concentration, and pH.¹⁵ The high negative ζ potentials for nonpolar organic liquids dispersed in brine solutions have been primarily attributed to the accumulation of hydroxyl ions at the oil/brine interfaces.^{14,15} In contrast to the simple organic liquids, crude oil contains surfactant-like components, such as

asphaltenes and resins, and these components add to the complexity of the charge distribution at oil/brine interfaces. Below, we report the ζ potentials of oil/brine interfaces in terms of ion type, brine concentration, and temperature. Figure 8 shows that, at low brine concentrations, the ζ potential reaches a value close to -100 mV for both oils 1 and 2. The ζ potential for oil/brine systems reported in previous work typically ranges from -50 to 0 mV,^{18–21} although higher negative values were also observed.^{22,23} The large negative value observed in our study is probably caused by the accumulation of hydroxyl ions and polar molecules of crude oil, such as asphaltene and resin, at the interface. As the salt concentration increases, the ζ potential generally becomes less negative, with a value close to zero at the highest salt concentrations examined. This observation is consistent with those from previous work,^{18,20,22} and the trend may be due to the adsorption of Na^+ and Mg^{2+} ions to the interface to compensate for the negative charges. Figure 8 also shows that a higher temperature generally has no discernible effect on the ζ potential of oil 1/brine systems and that some slight temperature effects were observed in oil 2/brine systems only at very low salt concentrations.

3.3. Discussion. Several previous studies have attempted to establish a direct link between the ζ potential and contact angle.^{5,20} However, we will show in the following discussion that there are complexities in establishing such a relationship. We begin by briefly reviewing the structure of the oil/brine/rock contact region. A thin brine film may exist between the rock and oil droplet submerged in a brine solution.¹ The brine phase of the oil/brine/rock contact region can be roughly divided into three regions, as illustrated in Figure 9. Region 1 is the thin brine film located between the oil droplet and rock. Region 2 is the contact region between the brine and rock far away from the thin brine film. Region 3 is the contact region between the brine and oil meniscus. The angle of the macroscopic meniscus when extrapolated to zero thickness of the brine film is normally defined as the contact angle.

The contact angle is influenced by the thickness of the thin brine film, which is affected by the charges at rock/brine and

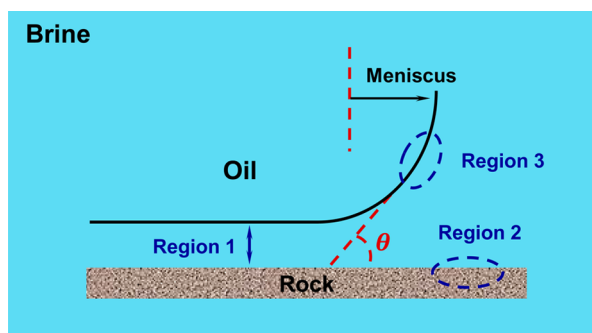


Figure 9. Structure of the oil/brine/rock contact region.

oil/brine interfaces of the thin film.¹ If the charges favor the repulsion of these two interfaces, the thin film may become thicker and the rock surface may become more water-wet, corresponding to a smaller contact angle. Similarly, a thinner film indicates a more oil-wet surface and, thus, a larger contact angle. The charges at the rock/brine and oil/brine interfaces of the thin brine film, however, are difficult to determine experimentally. The ζ potential for oil/brine and rock/brine suspensions do not necessarily represent the charges at the oil/brine and brine/rock interfaces of the thin brine film; they simply represent the corresponding interfacial charges far away from the thin film (i.e., regions 2 and 3). The results from the molecular dynamics simulations by Jiménez-Ángeles and Firoozabadi provide a more complex relationship between the contact angle and film thickness in crude oils than in simple model oils, such as normal alkanes.²⁴ When crude oils contain polar components, their polar head may have an affinity for the thin film, and the polar components in crude oils may influence the thin liquid composition differently compared to simple model oils. Accordingly, the properties of brine/rock and oil/brine interfaces far away from the thin film may be very different from the properties of the interfaces of the thin film.²⁴ Consequently, the ζ potential determined from oil/brine and rock/brine may not account for the critical role of the brine thin film in determining the contact angle. We have already started a comprehensive investigation along the lines of Jiménez-Ángeles and Firoozabadi,^{13,24} and results will be reported in future publications.

4. CONCLUSION

Raising the temperature from 25 to 50 °C has no noticeable effect on an oil sample, from a sandstone reservoir, on mica and quartz in both NaCl and MgCl₂ solutions over a concentration range of 0–3 M. The contact angles of an oil sample from a carbonate reservoir on calcite in NaCl and MgCl₂ solutions of 0–1 M decrease as the temperature increases from 25 to 65 °C, indicating a more water-wetting status. This temperature effect also strongly depends upon the brine type and salt concentration. In NaCl solution, the temperature effect is more pronounced at higher salt concentrations, whereas in MgCl₂ solution, the temperature effect has little dependence upon the salt concentration.

To shed light on the contact angle measurements, the ζ potential of rock/brine and oil/brine interfaces was examined in terms of the brine type, brine concentration, and temperature. The temperature change has little effect on the ζ potential of rock/brine interfaces. An elevated temperature generally has no appreciable effect on the ζ potential of the interface between the brine and oil from the sandstone

reservoir. The ζ potential of the interface between the brine and oil from the carbonate reservoir becomes less negative at a higher temperature only at very low salt concentrations. The measurements of the ζ potential of oil/brine and rock/brine interfaces and our discussions suggest that a direct link between the ζ potential and contact angle may not be established as a result of the overlook of the ζ potentials at the thin-film interfaces.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.energyfuels.7b00370.

Effect of aging time on the ζ potential of rock/brine (Figure 1) and oil/brine (Figure 2) interfaces (PDF)

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Notes

The authors declare no competing financial interest.

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