

The Effect of Water Salinity on Silica Dissolution Rate and Subsequent Formation Damage during Chemical EOR Process

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Abstract

During chemical EOR process, silicate scale has significant impact on the well productivity, rod pumps and other surface facility. The formation of silicate scale is a complex process involving silica dissolution, polymerization and subsequent precipitations. This paper presents the results of static and dynamic experiments that describe the effect of injection water salinity on silica dissolution rate and subsequent impact on formation permeability. Various synthetic brine salinities were utilized to determine the change in the silica dissolution rate using sandstone core samples. Results from static experiments indicated that 6.5% of the original silica was dissolved with the highest brine salinity of 60,000 ppm. Additional results demonstrated that the silica dissolution ratio has a significant effect on the initial core permeability. Using 60,000 ppm brine and 2.5% alkali, the initial permeability was reduced from 25.3 mD to 20.3 mD. The corresponding permeability reduction ratio for this case was 19.76%, which is equivalent to silica dissolution ratio of 15.99%. Finally, it is found that the brine salinity and pH has a pronounced impact on silica dissolution rate during chemical EOR process.

Keywords: Water injection; Silica dissolution; Silicate scale; Brine

Introduction

Traditionally, oil production strategies have followed primary depletion, secondary recovery and tertiary recovery processes. Secondary recovery methods are processes in which oil is subjected to immiscible displacement with injected fluids such as water or gas. One of the secondary recovery methods is to inject water into either oil formation or reservoir to contribute to the production of oil through oil displacement and pressure maintenance [1]. Water injection represents a reliable and economic approach to increase the percentage of oil recovery after primary production. In most cases, oil reservoirs suitable for water injection projects have been produced for several years. Usually, it takes time to inject sufficient water to fill enough of the void spaces to displace some of the remaining oil. The flood will probably have maximum recoveries during the second, third and fourth years after injection of water commenced [2]. Thus, an adequate strategy that accounts for all key factors affecting water injection project must be extensively planned. Water source and quality is one of the important issues controlling the performance of the project and should be given a priority in designing such project. Improper or inadequate water treatment can cause problems as undesirable precipitates or scale that may lead to unnecessary delay and economic failure [2]. In recent years, most waterflood projects have used saline or other nonpotable water, including formation water and sea water for offshore oil fields [2]. Sea water is injected under high pressure into the reservoir via injection wells to displace some of the remaining oil through the formation into nearby production wells. As the injection water travels through the sandstone formation, quartz silica is motivated to dissolve within the water. The interaction of the injected sea water with the formation water would decrease the pH of the mixed waters [3]. This reduction in the pH can significantly reduce the solubility of the dissolved monomeric silica [4]. At pH levels below 10.5, silica begins to polymerize and form colloidal silica. The hydroxide ions present in the mixed fluids catalyze the polymerization of silica [5]. Silica deposition is one of the major scale problems encountered during water injection process. It is a subsequent product generated after the polymerization of the dissolved monomeric silica within the mixed waters. The silicate scale depends on the pH and silica dissolution rate during the

interaction of the injected water with reservoir brine [5]. When the water is just injected into a sandstone reservoir, the high pH water dissolves quartz as monomeric silica ($\text{Si}(\text{OH})_3\text{O}^-\text{Na}^+$). High pH water tends to increase the rate of dissolution while keeping the monomeric silica soluble and under saturated. As the injection water flow through the formation, the high pH of the injection water is partially neutralized by the formation water. The low pH would lead the soluble silica to polymerize and forms colloidal silica nanoparticles. Colloidal silica forms when the solubility level of monomeric silica is exceeded in a supersaturated condition [5]. Additionally, below pH of 8.5, a favorable amorphous silica scaling is formed and results in polymerization and subsequent insolubility of silica [6]. Quartz dissolution from sandstone reservoirs can cause a significant dissolution of the silicate minerals, sandstone weight loss, propagation of significant concentrations of water-soluble silicates and hydroxide ion consumption. Further, the silicate scale can also precipitate in the production facilities and can reduce the well productively by reducing formation permeability at the vicinity of the production well [7,8]. An oil field operated in southern Alberta is a good example to experience the impact of silicate scale. The silicate scale had damaged the production wells and high concentration of silica inhibitor was used to reduce the impact of silicate scales [4]. Hence, many studies have been conducted recently to characterize and understand the key factors that affecting the mechanism of silicate scale during different conditions. It has been found that silica dissolution rate, silica polymerization and silicate scales are dependent on several factors; however the important three factors that have remarkable

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Received October 06, 2014; Accepted February 28, 2015; Published March 05, 2015

Citation: Elraies KA, Basbar AEA (2015) The Effect of Water Salinity on Silica Dissolution Rate and Subsequent Formation Damage during Chemical EOR Process. J Pet Environ Biotechnol 6: 209. doi:10.4172/2157-7463.1000209

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effect are pH, ion concentrations, and temperature [4-5,8-10]. This paper investigates the effect of the salinity and pH of injection water on silica dissolution rate. These are the same factors that control rates of silica deposition. Increased rate of silica dissolution therefore implies increased risk of silica scaling. Hence, series of static experiments were conducted to study the effect of different brine salinity on silica dissolution ratio. The effect of silica dissolution on core permeability was also investigated using dynamic test. The silica concentration before and after saturating with sandstone core sample was measured using X-RAY Fluorescence (XRF).

Materials and Methods

Materials description

Berea sandstone core was widely recognized by the petroleum industry as the best stone represent the reservoir. Hence, Berea sandstone core samples were used to conduct the static and dynamic experiments. The core was supplied by Cleveland Quarries TM. The alkali used in this study was sodium hydroxide (NaOH) with a purity of 99.0% and supplied by R&M TM. For polymeric inhibitor, PolyAcrylic Acid ($C_3H_4O_2$) with molecular weight of 1,800 was supplied by ALDRICH TM. Only sodium chloride (NaCl) was used for the preparation of different synthetic brine salinities. The sodium Chloride NaCl with 99.5% purity was supplied by MerckTM.

Apparatus and methods

Static and dynamic experiments were conducted to investigate the effect of brine salinity, pH and inhibitor on the silica dissolution rate. For the static test, different brine solutions were prepared using different brine salinities. The pH of these solutions was kept constant at seven as a worst-case scenario [6]. Consequently, 10 g of crushed quartz sand was added to 50 ml of each brine solution and the samples were then kept at room temperature for three days to achieve equilibrium condition. After equilibrium, all solutions were dried in oven and the silica concentration (SiO_2) before and after equilibrium was determined using X-RAY Fluorescence (XRF). This procedure was also repeated for the same brine salinities but with the presence of 2.5% of sodium carbonate as an alkaline agent. Additional experiments were also performed to investigate the effect of PloyAcrylic Acid (PAA) in inhibiting silica dissolution during static test. Equation 1 was used to measure the silica dissolution ratio as a function of SiO_2 concentration before and after saturating with brine solution.

$$SDR = \frac{[SiO_2]_{initial} - [SiO_2]_{blank}}{[SiO_2]_{initial}} * 100 \quad (1)$$

Where SDR is the silica dissolution ratio, $[SiO_2]_{initial}$ is the silica concentration before saturating the crushed core with brine and $[SiO_2]_{blank}$ is the silica concentration after the saturation.

Series of dynamic experiments were performed to investigate the effect of silica dissolution on permeability change during the water injection process. Different brine salinities with 2.5% alkali were utilized for each run. In carrying out the dynamic test, the initial permeability (K_1) of the core sample was determined by injecting pure distilled water. After determining the original permeability, synthetic brine with a particular salinity and 2.5% alkali was injected and the core permeability (K_2) was recorded for estimating the permeability reduction ratio (PRR). Another test was conducted to study the effect of silica dissolution in the presence of PloyAcrylic Acid (PAA) as inhibitor. Similar injection procedure was used; however, the last step

involves injecting synthetic brine with a particular salinity and 2.5% alkali with the presence of 250 mg/l of PAA. The core permeability during the last stage (K_3) was also measured for determining the permeability enhancement ratio (PER).

All dynamic tests were conducted using Bench Top Liquid Permeability System (BPS-805). All measurements were conducted at 1500 psi and ambient temperature. The flow rate was kept constant at 0.1cc/min for all runs and permeability for each stage was auto calculated by the built-in software in the Bench Top System. The permeability reduction ratio was calculated in the absence of the inhibitor using Equation 2.

$$PRR = \frac{K_1 - K_2}{K_1} \quad (2)$$

Where PRR is the permeability reduction ratio, K_1 and K_2 are the initial permeability using distilled water and brine permeability respectively.

The permeability enhancement ratio was calculated in the presence of the silica dissolution inhibitor using Equation 3.

$$PER = \frac{K_3 - K_1}{K_1} \quad (3)$$

Where PER is the permeability enhancement ratio and K_3 is the brine permeability in the presence of PAA.

Results and Discussion

Basic core morphology

The basic core mineralogy of the core sample used in this study was first identified by X-RAY diffraction (XRD and confirmed with X-RAY Fluorescence (XRF). Based on XRD results shown in Figure 1, the quartz or silica (SiO_2) is the main ingredient of the core followed by Kaolinite ($Al_2Si_2O_5(OH)_4$) and hematite (Fe_2O_3). It is clearly shown that silica has a significant quantity of the overall component. This is shown clearly at different peaks and peak 3 is the significant among others. The next highest peaks are 11 and 12 which refers to hematite and kaolinite, respectively. The kaolinite is partly filling the pores, and it occurs as face to face stacks of pseudo-hexagonal plates or books, however the hematite or the iron-oxide is one of the cementing factors [11].

X ray fluorescence (XRF) was then used to confirm the results obtained from XRD and also to determine the concentration of each component present in the core sample. Table 1 presents the concentration of each component. It was confirmed that silica (SiO_2) is the main component present in the core sample. The concentration of silica was found to be 94.4%, followed by aluminum oxide Al_2O_3 with 3.11%. The aluminum oxide represents a part of the kaolinite, which is a mixture of the aluminum oxide and silica hydroxide. The contribution of the rest of minerals is less than 1%.

Static experimental results

The effect of brine salinity and pH on silica dissolution: Two experiments were performed to investigate the effect of salinity and pH on silica dissolution using various brine salinities ranging from 5000 ppm to 60,000 ppm. In the first test, the pH of all solutions was adjusted to seven while in the second test; the pH was varied due to the presence of salinity and the addition of 2.5% of alkali. The silica concentration before and after saturating the crushed core with the solution was determined using X-RAY Fluorescence. The results obtained from

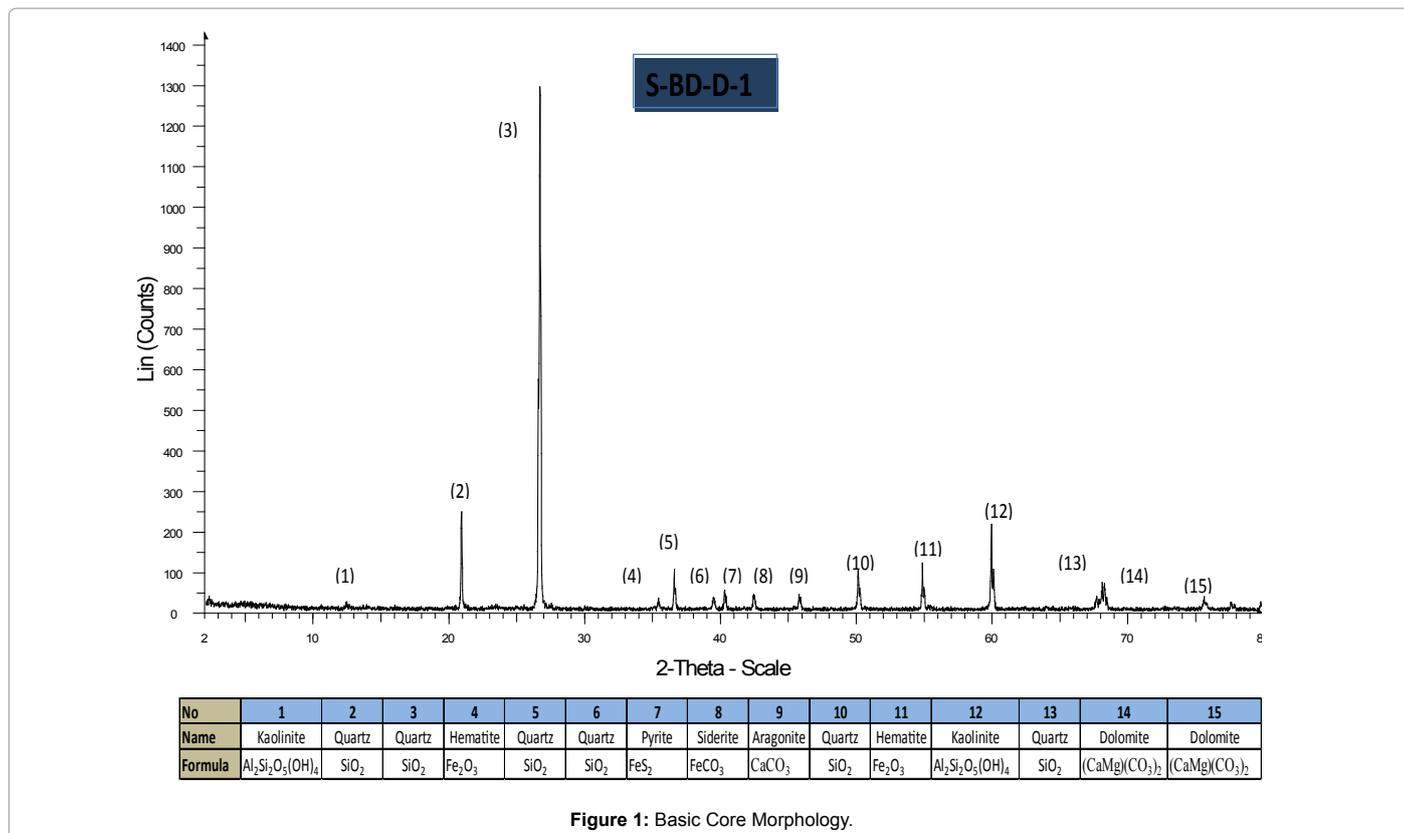


Figure 1: Basic Core Morphology.

| Minerals name | Chemical symbol | Concentration, % |
|--------------------|--------------------------------|------------------|
| Silica | SiO ₂ | 94.4000 |
| Aluminum oxide | Al ₂ O ₃ | 3.1100 |
| Potassium oxide | K ₂ O | 0.9680 |
| Iron (III) oxide | Fe ₂ O ₃ | 0.6304 |
| Sodium oxide | Na ₂ O | 0.2280 |
| Magnesium oxide | MgO | 0.1930 |
| Tedium oxide | TiO ₂ | 0.1610 |
| Calcium oxide | CaO | 0.1550 |
| Phosphate (oxide). | P ₂ O ₅ | 0.0190 |
| Zirconium dioxide | ZrO ₂ | 0.1286 |
| Copper(II) oxide | CuO | 0.0020 |
| Rubidium oxide | Rb ₂ O | 0.0030 |
| Zinc oxide | ZnO | 0.0020 |
| Total | | 100.0000 |

Table 1: Concentrations of minerals present in the core sample.

these experiments are tabulated in Table 2 and illustrated graphically in Figure 2.

Figure 2 shows the silica dissolution ratio versus brine salinity for both constant and variable pH solutions. As shown in Figure 2, the silica dissolution was significantly affected by the salinity and pH of the brine. It was also observed that the silica dissolution rate increases further as the salinity and pH of the brine was increased. For instance, with 60,000 ppm brine and seven pH solution, only 6.45% of the original silica was dissolved due to hydrolyze of the silica in the presence of sodium in the brine. The sodium acts as catalyst for

| Sample ID | Brine Salinity ppm | pH | SC % | SDR % |
|--|--------------------|---------------|-------|-------|
| S-BD-F-1 | - | - | 94.40 | - |
| Constant pH- In the absent of Alkali | | | | |
| S-AD-F-2 | 60,000 | Adjusted to 7 | 88.31 | 6.45 |
| S-AD-F-4 | 40,000 | | 89.18 | 5.53 |
| S-AD-F-6 | 25,000 | | 90.18 | 4.47 |
| S-AD-F-8 | 10,000 | | 91.53 | 3.04 |
| S-AD-F-10 | 5,000 | | 92.37 | 2.15 |
| Variable pH – in the presence of 2.5% Alkali | | | | |
| S-AD-F-12 | 60,000 | 13.20 | 79.31 | 15.99 |
| S-AD-F-14 | 40,000 | 13.01 | 80.08 | 15.17 |
| S-AD-F-16 | 25,000 | 12.85 | 84.18 | 10.83 |
| S-AD-F-18 | 10,000 | 12.63 | 88.53 | 6.22 |
| S-AD-F-20 | 5,000 | 12.34 | 90.32 | 4.32 |

Table 2: The effect of different brine salinities on silica concentration (SC) and silica dissolution ratio (SDR) before and after saturation.

dissolution and would chemisorb to increase the coordination number and weaken oxygen bonding [12]. On the other hand, 15.99% of the silica was dissolved when the pH was increased from 7 to 13.20 in the case of 60,000 ppm. Beside the influence of sodium within the solution, the high pH ionizes the dissolved silica which reduces the soluble silica concentration in the solution. This makes the system to re-equilibrate again by dissolving additional silica from the crushed core.

The effect of the inhibitor was investigated using 250 mg/l of PAA and various brine salinities with 2.5% alkali concentration. Thereafter, the results obtained from this experiment were compared with the results derived in the absence of inhibitor as presented in Figure 3. The use of 250 mg/l of PAA shows a significant reduction on the

| Sample ID | Salinity ppm | Liquid Permeability K | | | *PRR % | *PER % | SDR* % |
|-----------|--------------|-----------------------|-------|-------------|--------|--------|--------|
| | | Distilled water | Brine | Brine + PAA | | | |
| | | md | md | md | | | |
| D-AD-K-5 | 60,000 | 25.3 | 20.3 | | 19.76 | | 15.99 |
| D-AD-K-4 | 40,000 | 23.5 | 19.5 | | 17.02 | | 15.17 |
| D-AD-K-3 | 25,000 | 21.85 | 18.65 | | 14.65 | | 10.83 |
| D-AD-K-2 | 10,000 | 20.1 | 17.72 | | 11.84 | | 6.22 |
| D-AD-K-1 | 5,000 | 19.12 | 17.17 | | 10.17 | | 4.32 |
| D-I-K-10 | 60,000 | 77.5 | 70.02 | 96.65 | | 24.71 | 5.30 |
| D-I-K-9 | 40,000 | 61.71 | 56.01 | 75.78 | | 22.80 | 5.20 |
| D-I-K-8 | 25,000 | 51.6 | 48.08 | 59.52 | | 15.35 | 3.81 |
| D-I-K-7 | 10,000 | 39.42 | 37.91 | 42.62 | | 8.12 | 2.24 |
| D-I-K-6 | 5,000 | 31.2 | 30.28 | 33.04 | | 5.90 | 1.59 |

*PRR obtained from Equation 1, *PER obtained from Equation 2, *SDR obtained from static experiments

Table 3: Effect of different brine salinity on core permeability.

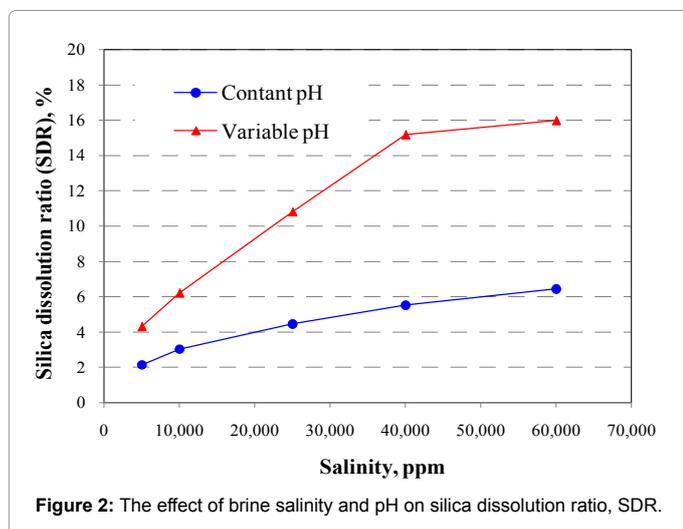


Figure 2: The effect of brine salinity and pH on silica dissolution ratio, SDR.

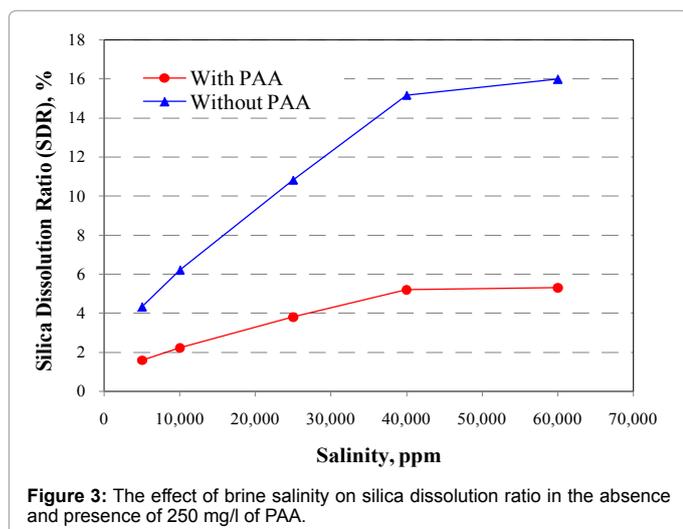


Figure 3: The effect of brine salinity on silica dissolution ratio in the absence and presence of 250 mg/l of PAA.

silica dissolution rate. At 60,000 ppm brine salinity, the dissolved silica decreased from 15.99% to 5.3%, while in case of lower salinity (5,000 ppm), the silica dissolution ratio was reduced from 4.3% to 1.6%. The reduction of the silica dissolution ratio can mainly be attributed to two reasons. The first reason is the reduction in the pH, which keeps

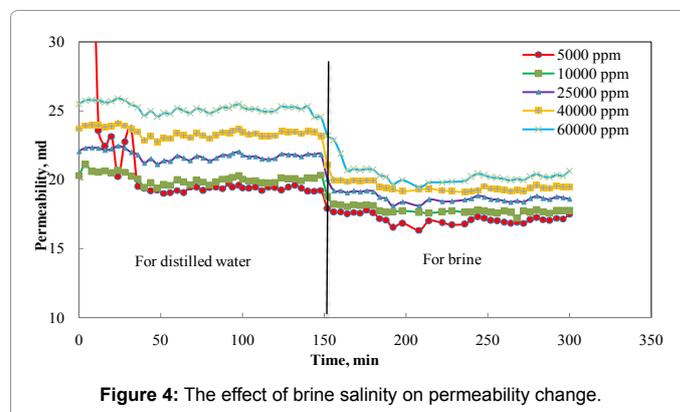


Figure 4: The effect of brine salinity on permeability change.

the solution in supersaturated conditions. The second reason is the reaction of the cationic and ionic molecules of the PAA with the soluble silica. This reaction or the trapping of the cationic polymers with the soluble silica leads to reduction of dissolving silica [6] (Figure 3).

Dynamic experimental results

The effect of brine salinity and pH on permeability reduction:

The aim of this test was to investigate the impact of salinity and pH on the permeability reduction caused by silica dissolution using Berea sandstone core sample. The effect of various brine salinities with 2.5% alkali was investigated in the absence and presence of 250 mg/l of PAA. A similar injection strategy was used for all experiments and the permeability during each step was recorded for comparison purposes. It should be noted that all permeability values reported in this paper are measured at stabilized pressure difference. The stabilized differential pressure after silica dissolution indicates that no more reaction within the porous media and Darcy law is valid for permeability measurement. The results obtained from these experiments are tabulated in Table 3 and illustrated graphically in Figure 4.

Figure 4 presents the change in the permeability for different brine salinities over the injection time. The initial permeability was significantly affected by the silica dissolution ratio (SDR) for different brine salinities. More reduction in the permeability was observed as the salinity was increased. This corresponds to SDR results obtained from the static experiments. For instance, the initial core permeability was reduced from 19.12 mD to 17.17 mD due to the injection of 5,000 ppm brine. This leads to permeability reduction ratio (PRR) of 10.17%, which is equivalent to SDR of 4.32%. On the other hand, the initial

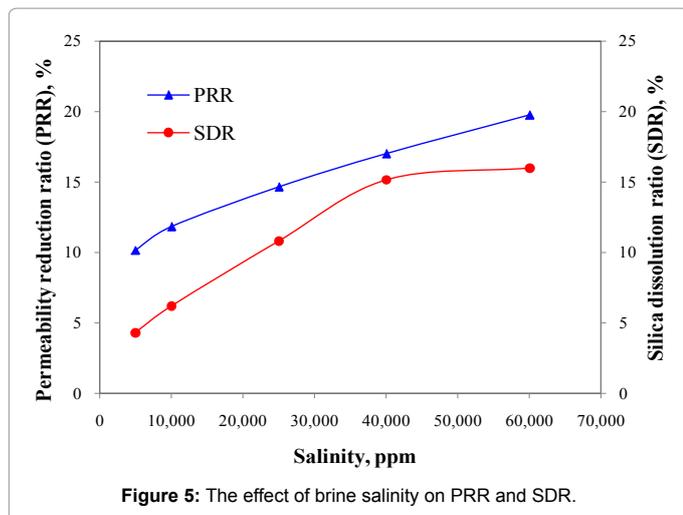


Figure 5: The effect of brine salinity on PRR and SDR.

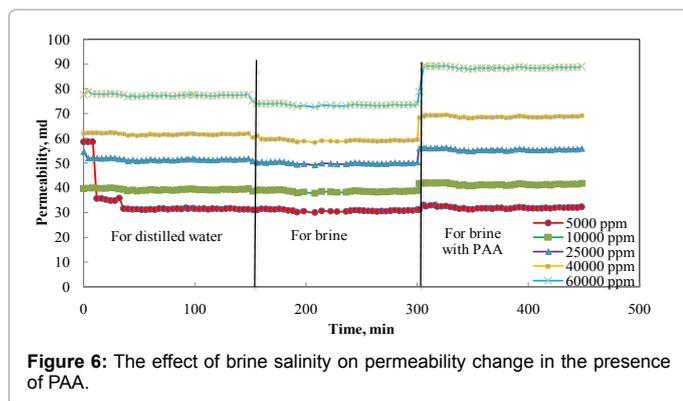


Figure 6: The effect of brine salinity on permeability change in the presence of PAA.

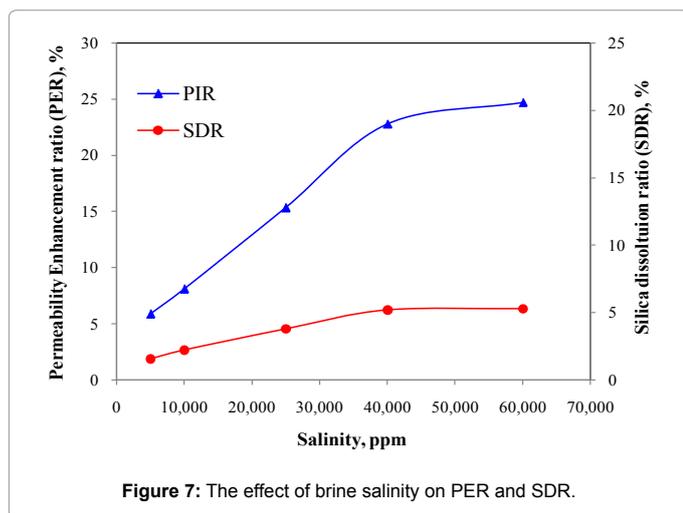


Figure 7: The effect of brine salinity on PER and SDR.

permeability was reduced from 25.3 mD to 20.3 mD when 60,000 ppm brine was injected through the core sample. The corresponding PRR of 19.76%, which is equivalent to SDR of 15.99%.

By referring to the results from both static and dynamic experiments, it is clear that the salinity and pH have significant effect in both silica dissolution and permeability reduction. Figure 5 shows the silica dissolution ratio (SDR) and the permeability reduction ratio

(PRR) as a function of brine salinity. It is observed that, the variation of the salinity with both silica dissolution and permeability reduction is generally in the same trend. It can be concluded that the increased rate of silica dissolution implies increased risk of formation damage.

Figure 6 shows the variation in the core permeability for various brine salinities in the presence of PAA. Obviously, the core permeability was significantly improved when 250 mg/l of PAA was introduced to the brine solution. Also, better enhancement in the permeability was observed at high brine salinity which had the highest SDR. The enhancement in the core permeability actually is attributed to the silica dissolution from the quartz. The possible reason is that when only brine is injected through the core sample, silica is separated from the core to be soluble within the solution. This soluble silica has a tendency of either precipitate to polymerize and form colloids that may remain suspended in the solution for long periods of time [13]. On the other hand, after injecting the brine with PAA, all silica might remain soluble in the solution and can be displaced with the solution, resulting in an increase in the core permeability. For instance, the initial core permeability was increased from 77.5 md to 96.65 md due to the injection of 60,000 ppm brine with 250 mg/l of PAA. The permeability enhancement ratio (PER) for this case is 24.17%, which is equivalent to SDR of 5.3%.

The use of the PAA with a concentration of 250 mg/l in both static and dynamic experiments shows significant effect in the core permeability. Figure 7 shows the silica dissolution ratio and the permeability enhancement ratio (PER) as a function of brine salinity.

Conclusion

Based on the findings and results obtained from the static experiments, it can be concluded that brine salinity has a significant effect on silica dissolution rate. It is apparent that an increase in brine salinity substantially increases silica dissolution rate which results in a more silica scale. The presence of sodium ions in the solution would catalyze the silica dissolution process, resulting in additional silica dissolution amount. However, the presence of alkali or high pH solution with the sodium ions would significantly increase the dissolution rate. The high pH ionizes the dissolved silica, making the system to re-equilibrate again by dissolving additional silica from the crushed core.

Results from the dynamic tests indicated that brine salinity and pH have a considerable effect on the original core permeability. The permeability reduction ratio (PRR) observed during the dynamic test was corresponding to the silica dissolution ratio (SDR) obtained from the static test. The higher SDR, the higher PRR is.

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