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Low-Sulfate Seawater Injection into Oil Reservoir to Avoid Scaling Problem

Amer Badr Bin Merdhah and Abu Azam Mohd Yassin
Faculty of Chemical and Natural Resources, Engineering Universiti Teknologi Malaysia,
81310 Skudai, Johor, Malaysia

Abstract: This study presents the results of laboratory experiments carried out to investigate the formation of calcium, strontium and barium sulfates from mixing Angsi seawater or low sulfate seawater with the following sulfate contents (75, 50, 25, 5 and 1%) and formation water contain high concentration of calcium, strontium and barium ions at various temperatures (40-90°C) and atmospheric pressure. The knowledge of solubility of common oil field scale formation and how their solubilities are affected by changes in salinity and temperatures is also studied. Results show a large of precipitation occurred in all jars containing seawater while the amount of precipitation decreased when the low sulfate seawater was used. At higher temperatures the mass of precipitation of CaSO_4 and SrSO_4 scales increases and the mass of precipitation of BaSO_4 scale decreases since the solubilities of CaSO_4 and SrSO_4 scales decreases and the solubility of BaSO_4 increases with increasing temperature. It can be concluded that even at sulfate content of 1% there may still be a scaling problem.

Key words: Scaling problems, low sulfate seawater, high salinity, high barium, temperature

INTRODUCTION

Secondary recovery is one of the activities used to improve oil recovery. We have several method used in secondary recovery. For example, water and gas flooding. The injection of water or gas into the oil-bearing reservoir is to increase the primary recovery factor and to maintain the reservoir pressure. In water flooding, the injected water will react with both the water already in the pore space of the rock (formation water) and with the mineral in the rock itself. This reaction will create scale formation.

Sulfate scale may result from changes in temperature and/or pressure while water flow from one location to another, but the major cause of sulfate scaling is the chemical incompatibility between the injected water, with high concentration of sulfate ion and formation waters, with high concentrations of calcium, barium and strontium ions.

Scale can occur at/or downstream of any point in the production system, at which super-saturation is generated. Super-saturation can be generated in single water by changing the pressure and temperature conditions or by mixing two incompatible waters. Changes in temperature, pressure, pH and $\text{CO}_2/\text{H}_2\text{S}$ partial pressure could also contribute in forming a scale (Mackay *et al.*, 2003; Moghadasi *et al.*, 2003a).

The objective of this work is to investigate the formation of calcium, strontium and barium sulfates from mixing Angsi seawater or low sulfate seawater and formation water at various temperatures and

concentrations. The knowledge of solubility of common oil field scale formation and how their solubilities were affected by changes in salinity and temperatures was also studied.

SCALE DEPOSITION

Scale deposition in surface and subsurface oil and gas production equipment has been recognized. Scale deposition is one of the most important and serious problems that inflict oil field water injection systems. Scale limits and sometimes blocks oil and gas production by plugging the oil-producing formation matrix or fractures and perforated intervals. It can also plug production lines and equipment and impair fluid flow. The consequence could be production-equipment failure, emergency shutdown, increased maintenance cost and overall decrease in production efficiency. The failure of these equipments could result in safety dangers. In case of water injection systems, scale could plug the pores of the formation and results in injectivity decline with time (Yuan and Todd, 1991; Bayona, 1993; Asghari and Kharrat, 1995; Andersen *et al.*, 2000; Paulo *et al.*, 2001; Voloshin *et al.*, 2003). Scale deposition can occur from one type of water because of super-saturation with scale-forming salts attributable to changes in the physical conditions under which the water exists. Scale also can deposit when two incompatible waters are mixed and super-saturation is reached (Todd and Yuan, 1992; Moghadasi *et al.*, 2003b, 2004).

SOURCE OF OIL FIELD SCALE

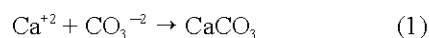
The chief source of oil field scale is mixing of incompatible waters. Two waters are called incompatible if they interact chemically and precipitate minerals when mixed. A typical example of incompatible waters are sea water with high concentration of SO_4^{-2} and low concentrations of Ca^{+2} , $\text{Ba}^{+2}/\text{Sr}^{+2}$ and formation waters with very low concentrations of SO_4^{-2} but high concentrations of Ca^{+2} , Ba^{+2} and Sr^{+2} . Mixing of these waters, therefore, causes precipitation of CaSO_4 , BaSO_4 and/or SrSO_4 . Field produced water (disposal water) can also be incompatible with seawater. In cases where disposal water is mixed with seawater for re-injection, scale deposition is possible (Bayona, 1993; Andersen *et al.*, 2000; Bedrikovitsky *et al.*, 2001; Stalker *et al.*, 2003; Paulo *et al.*, 2001).

During the production, the water is drained to the surface and suffers from significant pressure drop and temperature variations. The successive pressure drops lead to release of the carbon dioxide with an increase in pH value of the produced water and precipitation of calcium carbonate (Mackay, 2003).

COMMON OIL FIELD SCALES

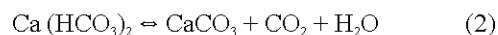
The most common scales encountered in oil field operations are sulfates such as calcium sulfate (anhydrite, gypsum), barium sulfate (barite) and strontium sulfate (celestite) and calcium carbonate. There follows a brief description of each scale.

Calcium carbonate scales: The water is drained to the surface and suffers from significant pressure drop and temperature variations during the production. The continuous pressure drops lead to degassing of the carbon dioxide with an increase in pH value of the produced water and precipitation of calcium carbonate (Mackay, 2003; Rousseau *et al.*, 2003). Calcium carbonate or calcite scale is frequently encountered in oil field operations. But the calcite has the greatest stability in oilfield circumstances, so it is the most common form of calcium carbonate encountered in oilfield production operation. Deposition of CaCO_3 scale results from precipitation of calcium carbonate according to the following equation:



As it will be seen later calcium carbonate scale can also be formed by combination of calcium and bicarbonate ions and this reaction is the major cause of calcium carbonate scale deposition in oilfield operations. This is

because only a small percentage of the bicarbonate ions dissociated at the pH values found in most injection waters to form H^+ and CO_3^{-2} (Moghadasi *et al.*, 2004). Moreover, carbonate scale formation occurs when connate water or aquifer water passes through the bubble point and carbon dioxide is evolved. As carbon dioxide is evolved, the solubility with respect to carbonate declines rapidly and forms a precipitate with divalent ions, such as iron and more commonly calcium, as outlined in the following equation (Mackay and Jordan, 2005):



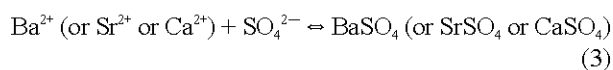
Calcium sulfate scales: Calcium sulfate scale poses a unique problem for the salts under consideration because it occurs with one of three different phases. Calcium sulfate exists in several crystalline forms. These include gypsum ($\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$) and anhydrite (CaSO_4). Gypsum, the most common scale former occurs at relatively low temperature. At higher temperature (above 100°C), the stable phase predicted is anhydrite (CaSO_4). However, hemi-hydrate has been known to form at 100 to 121°C , especially in non-turbulent systems and in high ionic strength brines (Moghadasi *et al.*, 2003a). These compounds may be stable depending on temperature and ionic strength and they have decreasing solubilities with increasing temperatures above 40°C .

Among various types of mineral scales, calcium sulfate is one of major scales in petroleum industry that can cause severe flow assurance and formation damage issues. Many parameters are affecting this problem. Temperature, pressure, fluid concentration, ratio of brine to hydrogen, fluid dynamic and type of porous media are among these parameters (Tahmasebi *et al.*, 2007).

Barium sulfate scale: The barium sulfate scaling is a chronic disaster in waterflood projects with incompatible injected and formation waters. This is usually due to precipitation of BaSO_4 from the mixture of both waters and consequent permeability reduction resulting in well productivity decrease (Bedrikovitsky *et al.*, 2006). Moreover, barium sulfate is the most insoluble scale that can be precipitated from oilfield waters. It forms a hard scale which is extremely difficult to remove. The solubility of barium sulfate is about a thousand times less than of calcium sulfate, at surface conditions. Furthermore, the solubility of barium sulfate goes up with increasing temperature, pressure and salt content of the brine. Thus prediction of barium sulfate scale is much easier than the others since a pressure, temperature or salt content drop will cause increased precipitation.

In the postseawater-breakthrough period, however, there is a much more serious problem of precipitation of

barium sulfate from an incompatibility between the formation water and seawater. BaSO₄ scale removal is particularly difficult. Thus, BaSO₄ scale treatment must focus mainly on its prevention through the use of scale-control chemicals. Thus, the severity of the scaling problem is determined both by the scaling rate and the efficiency of the chemical inhibitors (Mazzollini *et al.*, 1992). Moreover, the case where water injection (seawater, river, aquifer, or produced water) is used for pressure maintenance and sweep, the mixing of incompatible brines can lead to the formation of sulfate scales when the injection water contains sulfate ions (Mackay and Jordan, 2005).



Strontium sulfate scale: Strontium sulfate scale formation has become a growing concern in oil-production systems. Until recently, the appearance of strontium in oilfield scales has been primarily in the presence of barium sulfate scale. Almost pure SrSO₄ scale now is observed in several production wells around the world. The scale formation is primarily a result of subsurface commingling of waters, which results in water supersaturated in SrSO₄.

According to Lindlof and Stoffer (1983), strontium sulfate solubility is decreased by the common ion effect; the super-saturation becomes a disproportionately higher percentage of total strontium sulfates in the solution. The super-saturation represents the amount of strontium sulfate present in excess of the solubility and thus represents the amount available for precipitation from solution and possible scaling. The super-saturation exists in a metastable state and as such, the manner in which it exists in solution or comes out of solution by crystallization and precipitation is entirely unpredictable.

SOLUBILITY OF SCALES

Solubility is defined as the limiting amount of solute that can dissolve in a solvent under a given set of physical conditions. According to Oddo *et al.* (1991), calcium sulfate scale formation is somewhat dependent on temperature, but is typically precipitated because of a decrease in pressure or an increase in the relative concentrations of calcium or sulfate. CaSO₄ solubility is fairly independent of pH and hence, can readily precipitate in an acid environment.

Strontium sulfate solubilities may play a role in many disciplines of science and engineering. For example, strontium sulfate forms scale in oil and/or geothermal fields which are frequently accompanied by other sulfates of alkaline earth metals. Jacques and Bourland (1983)

described a solubility study of strontium sulfate in sodium chloride brine. Their study showed that the solubility of strontium sulfate increased with increasing ionic strength and decreased with increasing temperature.

Barium sulfate scale (barite) in oil fields can be precipitated easily on the basis of already available information relating to thermodynamic condition and the kinetics of precipitation (Mitchell *et al.*, 1980). Barium sulfate solubility increased with temperature increase, with increase ionic strength of brine and with pressure. Barium sulfate precipitation was affected most strongly by temperature (Moghadas *et al.*, 2003a).

Factors that affect scale precipitation, deposition and crystal growth can be summarized as: super-saturation, temperature, pressure, ionic strength, evaporation, contact time and pH. Effective scale control should be one of the primary objectives of any efficient water injection and normal production operation in oil and gas fields.

THE SCALING PROBLEM IN OIL FIELDS

Scaling deposition is one of the most important and serious problems which water injection systems are generally engaged in. Scale deposited in down-hole pumps, tubing, casing flow-lines, heater treaters, tanks and other production equipment and facilities.

Scale formation is a major problem in the oil industry. They may occur down-hole or in surface facilities. The formations of these scales plug production lines and equipment and impair fluid flow. Their consequence could be production-equipment failure, emergency shutdown, increased maintenance cost and an overall decrease in production efficiency. The failure of production equipment and instruments could result in safety hazards (Yeboah *et al.*, 1993).

According to Bertero *et al.* (1988), one of the problems encountered in water flooding projects is scale formation caused by chemical incompatibility between potential injection waters and reservoir brine. Chemical compatibility evaluation through laboratory experiments on cores at reservoir conditions is of limited value because only first-contact phenomena are reproduced. A scale problem will occur, if at a high water cut part of the water is present as free water. The rate of scale deposition will then be approximately proportional to the rate of free water production. Depending upon where the formation water becomes supersaturated, scale may be deposited in the flow line only, in both flow line and tubing and in some cases even in the perforations and in the formation near the wellbore.

The formation of inorganic mineral scale within onshore and offshore production facilities around the world is a relatively common problem. Scale can form

from a single produced connate or aquifer water due to changes in temperature and pressure, or when two incompatible waters mix. An example of the latter would be seawater support of a reservoir where the formation water is rich in cations (Ba, Sr and Ca) and the injection water is rich in anions (SO₄).

Oil field scales costs are high due to intense oil and gas production decline, frequently pulling of down-hole equipment for replacement, re-perforation of the producing intervals, re-drilling of plugged oil wells, stimulation of plugged oil-bearing formations and other remedial workovers through production and injection wells. As scale deposits around the well-bore, the porous media of formation becomes plugged and may be rendered impermeable to any fluids.

The production problems caused by mineral scale in oil production operations have long been known. Among the most onerous of all scaling problems is that of sulfate scales, particularly barium sulfate scale. This is a difficult scaling problem because of the low solubility of barium sulfate in most fluids and the commensurate low reactivity of most acids with barium sulfate scale. Deposition of barium sulfate into a continuous scale surface on production tubular exposes very little surface area for treatment by chemicals and therefore this scale is almost impossible to remove once it is deposited. The most popular approach to addressing the barium sulfate scale problem has been to retard or prevent the formation of this scale in the first place (McElhiney *et al.*, 2001).

Many case histories of oil well scaling by calcium carbonate, calcium sulfate, strontium sulfate and barium sulfate have been reported (Mitchell *et al.*, 1980; Lindof and Stoffer, 1983; Vetter *et al.*, 1987; Shuler *et al.*, 1991). Problems in connection to oil well scaling in the Russia where scale has seriously plugged wells and are similar to cases in North Sea fields have been reported (Mitchell *et al.*, 1980). Oilfields scale problems have occurred because of water flooding in Saudi oil fields, Algeria, Indonesia in south Sumatra oilfields and Egypt in el-Morgan oilfield where calcium and strontium sulfate scales have been found in surface and subsurface production equipment (El-Hattab, 1982).

WHERE DOES OILFIELD SCALE FORM?

The scaling reaction depends on there being adequate concentrations of sulfate ions in the injected seawater and barium, strontium and calcium divalent cations in the formation brine to generate sulfate scale or on there being enough bicarbonate and calcium ions to generate carbonate scale.

Therefore scale precipitation may occur wherever there is mixing of incompatible brines, or there are changes in the physical condition such as pressure decline. An overview of all the possible scale formation environments for seawater, aquifer, natural depletion and produced water re-injection is presented in Fig. 1 (Jordan and Mackay, 2005; Jordan *et al.*, 2006).

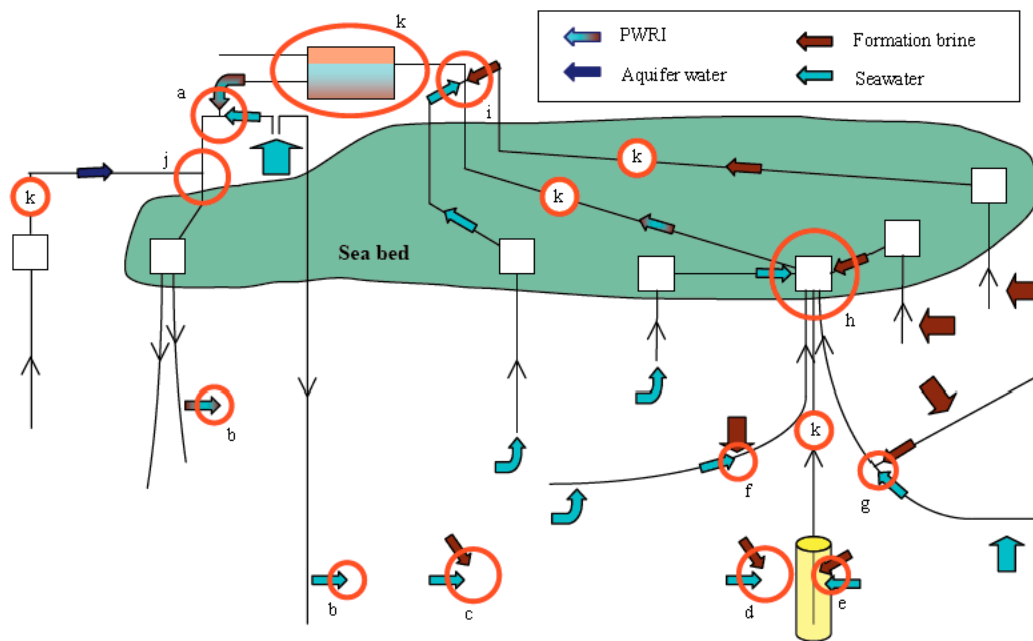
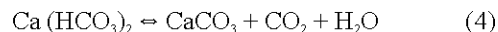


Fig. 1: Locations throughout the flow system where scale deposition may take place (Jordan and Mackay, 2005; Jordan *et al.*, 2006)

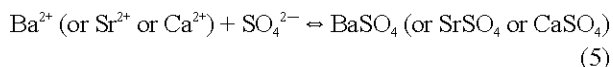
- Because injection, for example if seawater injection is supplement by Produced Water Re-Injection (PWRI).
- Around the injection well, as injection brine enters the reservoir, contacting formation brine.
- Deep in formation, due to displacement of formation brine by injected brine, or due to meeting flow paths.
- As injection brine and formation brine converge towards the production well, but beyond the radius of a squeeze treatment.
- As injection brine and formation brine converge towards the production well and within the radius of a squeeze treatment.
- In the completed interval of a production well, as one brine enters the completion, while other brine is following up the tubing from a lower section, or as fluid pressure decreases.
- At the junction of a multilateral well, where one branch is producing single brine and the other branch is producing incompatible brine.
- At a sub-sea manifold, where one well is producing single brine and another well is producing different brine.
- At the surface facilities, where one production stream is flowing one brine and another production stream is flowing another brine.
- During aquifer water production and processing for re-injection could lead to scale formation within self-scaling brine or mixing with incompatible formation brine.
- During pressure reduction and/or an increase in temperature within any down-hole tube or surface processing equipment, leading to the evolution of CO₂ and to the generation of carbonate and sulfide scale if the suitable ions are present. Temperature reductions could lead to the formation of halite scales if the brine was close to saturation under reservoir conditions.

Oilfield scales are inorganic crystalline deposits that form as a result of the precipitation of solids from brines present in the reservoir and production flow system. The precipitation of these solids occurs as the result of changes in the ionic composition, pH, pressure and temperature of the brine. There are three principal mechanisms by which scales form in both offshore and onshore oil field system (Mackay, 2005; Jordan and Mackay, 2005; Collins *et al.*, 2006):

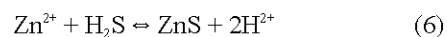
- Decrease in pressure and/or increase in temperature of a brine, goes to a reduction in the solubility of the salt (most commonly these lead to precipitation of carbonate scales, such as CaCO₃).



- Mixing of two incompatible brines (most commonly formation water rich in cations such as barium, calcium and/or strontium, mixing with sulfate rich seawater, goes to the precipitation of sulfate scales, such as BaSO₄).



Other fluid incompatibilities include sulfide scale where hydrogen sulfide gas mixes with iron, zinc or lead rich formation waters:



- Brine evaporation, resulting in salt concentration increasing above the solubility limit and goes to salt precipitation (as may occur in HP/HT gas wells where a dry gas stream may mix with a low rate brine stream resulting in dehydration and most commonly the precipitation of NaCl).

MATERIALS AND METHODS

The intention of this study was to show the effect of low sulfate seawater to avoid scaling problem when mixing seawater or low sulfate seawater with the following sulfate contents (75, 50, 25, 5 and 1%) and formation water contain high concentration of calcium, strontium and barium ions at various temperatures (40-90°C) and atmospheric pressure.

The ionic compositions of formation waters and Angsi seawater are given in Table 1. The formation water has calcium, strontium and barium ions and the sea water contains sulfate ions. It was clear that the mixing of these waters can lead to calcium, strontium and barium sulfate precipitation.

Table 1: The ionic compositions of formation and injection waters

Ionic	High salinity	High barium	Angsi seawater
	formation water	formation water	(Malaysia)
	----- (ppm) -----		
Sodium	52,132	42,707	10,805
Potassium	1,967	1,972	375
Magnesium	4,260	102	1,295
Calcium	30,000	780	429
Strontium	1,100	370	6.577
Barium	10	2,200	-
Chloride	146,385	67,713	19,307
Sulfate	108	5	2,750
Bicarbonate	350	2,140	158.80

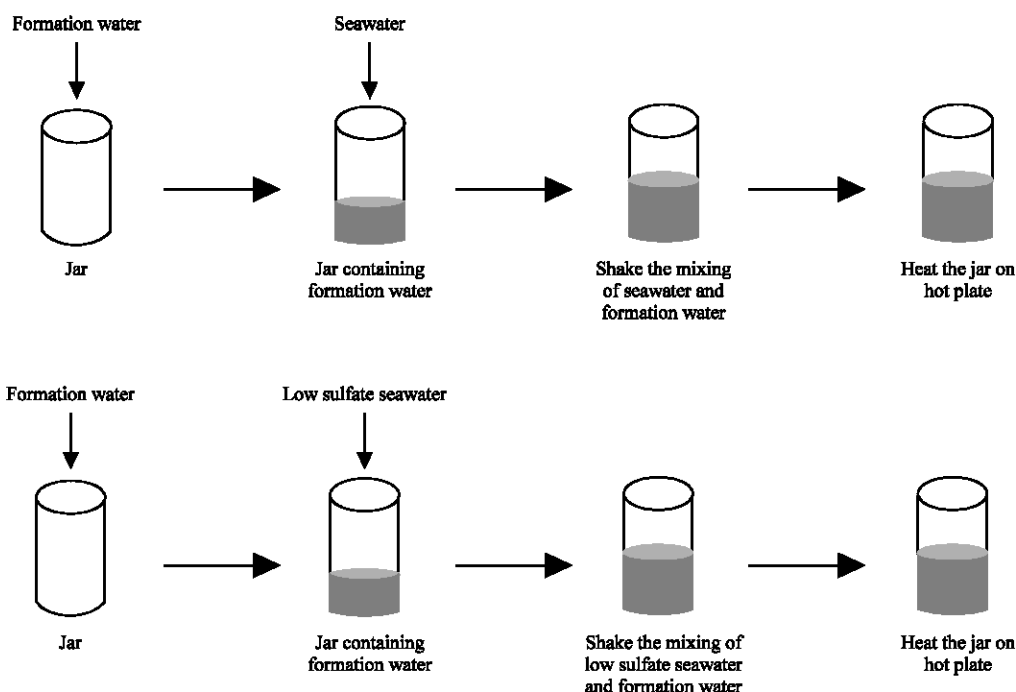


Fig. 2: Jar test using seawater and low sulfate seawater

The experimental procedures used in the determination of solubility of common oil field scales from mixing brines (formation water and Angsi seawater) are shown in Fig. 2:

- For each experiment of common oil field scales, 100 mL of each filtered opposite waters was poured simultaneously into a clean glass jar and shaken moderately.
- The mixture was heated on hot plate and the mixture was stirred by magnetic stirrer and after that the mixture was filtered through 0.45 μm filter paper.
- The crystals on the filter paper were dried in a humidity oven and the weight of dried crystal sample was measured by Electronic Top Pan Balance.
- After filtration, 5 mL of the filtrate was pipetted into a 50 mL volumetric flask and filled up with distilled water to 50 mL. This instantaneous dilution of the CaSO_4 , SrSO_4 and BaSO_4 containing brines just after filtration was performed in order to prevent CaSO_4 , SrSO_4 or BaSO_4 precipitation during the period between filtering and analytical determination of the Ca, Ba and Sr concentrations.
- The calcium, barium and strontium determinations were calibrated by measuring five standard solutions. Standard solutions were prepared from CaCl_2 , BaCl_2 and SrCl_2 solutions.

- Calcium, barium and strontium concentrations in the diluted filtrates were determined by atomic absorption spectrometry. After multiplying with the dilution factor, the exact concentrations of calcium, barium and strontium were computed.

RESULTS AND DISCUSSION

The main objective of this study to investigate the formation of calcium, strontium and barium sulfates from mixing seawater or low sulfate seawater with the following sulfate contents (75, 50, 25, 5 and 1%) and formation water contain high concentration of calcium, strontium and barium ions at various temperatures (40-90°C) and atmospheric pressure. In the following, the results for solubility of scale formation and low sulfate seawater experiments are discussed individually:

Solubility of scale formation experiments: The concentrations of calcium, barium and strontium in the diluted filtrates were determined by atomic absorption spectrometry. The solubilities of CaSO_4 , BaSO_4 and SrSO_4 at various temperatures (40-90°C) of this study were calculated. Graphical presentation is given in Fig. 3.

The expected trend in this temperature range is a decrease of CaSO_4 and SrSO_4 solubilities, but an increase of BaSO_4 solubility with increasing temperature.

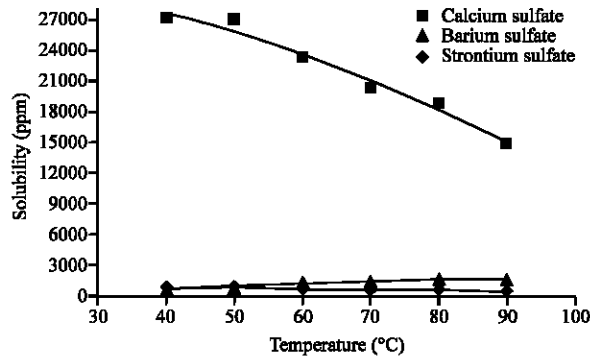


Fig. 3: Solubility of scale formation is dependent on temperature

A graphical presentation of the experimental results (Fig. 3) illustrates this trend in these experiments. The SO_4^- content of the sea water brine reacts with the barium ions of the formation water instantaneously, but it reacts of both calcium and strontium ions during heating.

The experimental results confirm the general trend in solubility dependencies for common oil field scales with temperature. They are obvious and similar to that observed in the earlier works (Jacques and Bourland, 1983; Lindlof and Stoffer, 1983; Oddo *et al.*, 1991; Moghadasi *et al.*, 2003a; Mackay, 2003; Rousseau *et al.*, 2003; Mackay and Jordan, 2005).

Low sulfate sea water experiments: The objective of this part of the investigation is to study the effect of using seawater and low sulfate seawater to avoid scaling problem during water injection.

In these experiments, the sea water and low sulfate seawater were mixed with formation water at the condition of high-salinity (high concentration of calcium and strontium) and high concentration of barium. Temperatures (40-90°C) effects are conducted to give insight into the nature of the scale.

Figure 4 and 5 show that the sulfate content in sea water play an important role in precipitation of sulfate scale. Generally, it could be concluded that the mass of precipitation increased when the sulfate content in sea water is high.

Moreover, when sulfate content in seawater is 100%, we can see that the mass of precipitation of CaSO_4 and SrSO_4 scales is around 630 mg at temperature 90°C and the mass of precipitation of BaSO_4 scale is 680 mg at temperature 40°C. After the sulfate content in seawater is reduced to 75% concentration, the mass of precipitation has decreased gradually. Similar results can be observed for 50, 25, 5 and 1% sulfate concentration in seawater.

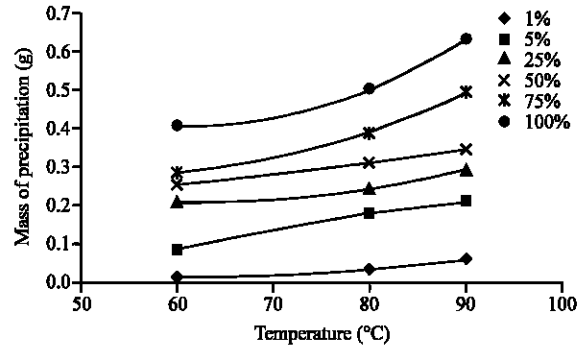


Fig. 4: Variation of mass of precipitation as a function of temperature showing the effect of sulfate concentration in seawater at high salinity

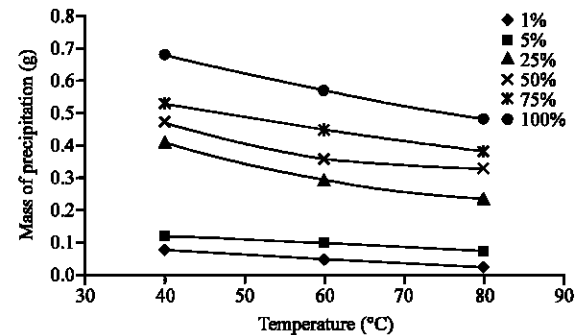


Fig. 5: Variation of mass of precipitation as a function of temperature showing the effect of sulfate concentration in seawater at high barium

From the Fig. 4 and 5, we can see that at 1% sulfate concentration, the precipitation still occurred, but the amount of precipitation is reduced by 90% at high salinity and 89% at high barium compared to 100% sulfate concentration. Therefore, we can conclude that even at the sulfate content in seawater of 1% of sulfate, there may still be a scaling problem.

Even though using low sulfate seawater could generate scaling potential, it is more acceptable compare to seawater in term of water injector. This is because low sulfate seawater only needs a few inhibitors to avoid scaling problem than seawater. Sulfate scale may result from changes in temperature and/or pressure while water flow from one place to another, but the major cause of sulfate scaling is the chemical incompatibility between the injected seawater, which originally contains high in sulfate ion and the formation, which originally contains high concentration of calcium, barium and/or strontium.

Generally, temperature plays a vital role in scale formation. From the Fig. 6 and 7 at higher temperatures

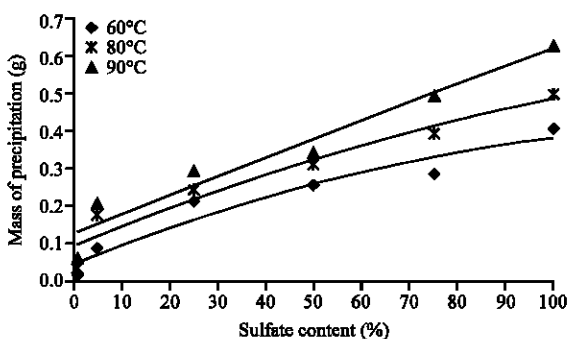


Fig. 6: Variation of mass of precipitation as a function of sulfate concentration in seawater showing the effect of temperature at high salinity

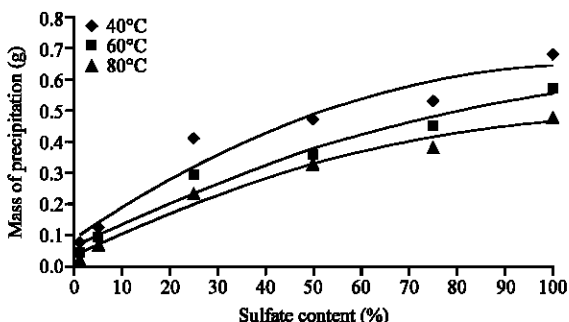


Fig. 7: Variation of mass of precipitation as a function of sulfate concentration in seawater showing the effect of temperature at high barium

the mass of precipitation of CaSO_4 and SrSO_4 scales increases and the mass of precipitation of BaSO_4 scale decreases since the solubilities of CaSO_4 and SrSO_4 scales decreases and the solubility of BaSO_4 increases with increasing temperature.

CONCLUSIONS

The work carried out in this study focused on the effect of low sulfate seawater to avoid scaling problem when mixing seawater or low sulfate seawater with the following sulfate contents (75, 50, 25, 5 and 1%) and formation water contain high concentration of calcium, strontium and barium ions at various temperatures (40-90°C) and the knowledge of solubility of scale formation and how its solubilities are affected by changes in salinity and temperature. Based on the results obtained from these studies, the following conclusions can be made:

- The experimental results confirm the general trend in solubility dependencies for common oil field scales

at various temperatures (40 to 90°C) and 1 atm. A temperature rise from 40 to 90°C causes an increase in BaSO_4 solubility, but a decrease in CaSO_4 and SrSO_4 solubilities.

- As the sulfate in seawater was increased, sulfate scale deposition increases. Even at sulfate content in seawater as low as 1%, sulfate scale is still being formed.
- At higher temperatures the mass of precipitation of CaSO_4 and SrSO_4 scales increases and the mass of precipitation of BaSO_4 scale decreases since the solubilities of CaSO_4 and SrSO_4 scales decreases and the solubility of BaSO_4 increases with increasing temperature.
- Low sulfate seawater can be used as water injector but it needs some inhibitor to avoid formation of scale.

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