



Journal of Applied Sciences

ISSN 1812-5654

science
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Barium Sulfate Scale Formation in Oil Reservoir During Water Injection at High-Barium Formation Water

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Abstract: This study presents the results of laboratory experiments carried out to investigate the formation of barium sulfate in sandstone cores from mixing injected sea water and formation water contain high concentration of barium at various temperatures (50 and 80°C) and differential pressures (100, 150 and 200 psig). The morphology of scaling crystals as shown by Scanning Electron Microscopy (SEM) is presented. Results show a large extent of permeability damage caused by barium sulfate deposits on the rock pore surface. The rock permeability decline indicates the influence of the concentration of barium ions.

Key words: Scale deposition, solubility of scale, concentration of barium ions, temperature, pressure

INTRODUCTION

The injection of seawater into oilfield reservoirs to maintain reservoir pressure and improve secondary recovery is a well-established, mature, operation. Moreover, the degree of risk posed by deposition of mineral scales to the injection and production wells during such operations has been much studied. Scale deposition is one of the most serious oil field problems that inflict water injection systems primarily when two incompatible waters are involved. Two waters are called incompatible if they interact chemically and precipitate minerals when mixed. Typical examples are sea water, with high concentration of SO_4^{-2} and formation waters, with high concentrations of Ca^{+2} , Ba^{+2} and Sr^{+2} . Mixing of these waters, therefore, could cause precipitation of CaSO_4 , BaSO_4 and/or SrSO_4 .

The most common oil field scales are listed in Table 1, along with the primary variables that affect their solubility (Moghadasi *et al.*, 2003a). These scales are sulfates such as calcium sulfate (anhydrite, gypsum), barium sulfate (barite) and strontium sulfate (celestite) and calcium carbonate. Other less common scales have also been reported such as iron oxides, iron sulfides and iron carbonate. Lead and zinc sulfide scale has recently become a concern in a number of North Sea oil and gas fields (Collins and Jordan, 2001).

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

Table 1: Most common oilfield scales

Name	Chemical formula	Primary variables
Calcium carbonate	CaCO_3	Partial pressure of CO_2 , temperature, total dissolved salts, pH
Calcium sulfate		
Gypsum	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	Temperature, total dissolved salts, pressure
Hemihydrate	$\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$	
Anhydrite	CaSO_4	
Barium Sulfate	BaSO_4	Temperature, pressure
Strontium Sulfate	SrSO_4	Temperature, pressure, total dissolved salts
Iron compounds		
Ferrous Carbonate	FeCO_3	Corrosion, dissolved gases, pH
Ferrous Sulfide	FeS	
Ferrous Hydroxide	Fe(OH)_2	
Ferrous Hydroxide	Fe(OH)_3	

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. Scale also deposited in down-hole pumps, tubing, casing flow-lines, heater treaters, tanks and other production equipment and facilities. The consequence could be production-equipment failure, emergency shutdown, increased maintenance cost and overall decrease in production efficiency. In case of water injection systems, scale could plug the pores of the formation and results in injectivity decline with time (Todd and Yuan, 1992; Bayona, 1993; Asghari and Kharrat, 1995; Andersen *et al.*, 2000; Paulo *et al.*, 2001; Voloshin *et al.*, 2003). Scale also can deposit when two incompatible waters are mixed and super-saturation is reached (Nassivera and Essel, 1979; Read and Ringen, 1982; Vetter *et al.*, 1982; Todd and Yuan, 1992; Moghadasi *et al.*, 2003b, 2004).

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} , Ba^{+2}/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).

Scale formation can be predicted by laboratory experiments. Several experimental studies have been conducted to determine the scaling potential in different oil-fields is listed in Table 2-4. In experimental studies, the extent of permeability damage caused by flowing precipitates in cores and sand packs was investigated. Several investigators tested incompatible waters from

Table 2: Summary of previous experimental studies

Rock	Scale	Concentration		Temperature (°C)	Pressure (Psig)	Flow rate	References
		Sea water (ppm)	Formation water (ppm)				
Sand pack	Barium sulfate and calcium carbonate	Table 4	Table 3	20-90	4-40	1:10	Mitchell <i>et al.</i> (1980)
Glass beads and in synthetic alumina cores	Barium, strontium, calcium, sulfates and calcium carbonate	Table 4	Table 3	20-70	1-22	2 mL min ⁻¹	Read and Ringen (1982)
Sand stone out crop (Brazil)	Barium and strontium sulfates	Table 4	Table 3	80	-	0.5-1.0 mL min ⁻¹	Bezerra <i>et al.</i> (1990)
Sand stone Clashach	Barium and strontium sulfates	Table 4	Table 3	20-70	1-2	7.5 mL min ⁻¹	Todd and Yuan (1992)
Berea Sand stone	Barium, strontium and calcium sulfates	Table 4	Table 3	21	14.7	-	McElhiney <i>et al.</i> (2001)
Sand pack and Glass beads	Barium, strontium and calcium sulfates	Table 4	Table 3	25-80	1-145	1-200 mL min ⁻¹	Moghadasi <i>et al.</i> (2002)

Table 3: Ions of formation water

Place	Mitchell <i>et al.</i> (1980)	Read and Ringen (1982)	Bezerra <i>et al.</i> (1990)	Todd and Yuan (1992)		McElhiney <i>et al.</i> (2001)		Moghadasi <i>et al.</i> (2002)
	Forties (North sea)		Namorado field (Brazil)	M (North sea)	ST (North sea)	West African F.W	F.W with out Ba	Mishrif in Iranian
	Ca	3110	1100	2760	2809	779	4000	4000
Ba	250	210	229	252	2180	240	0	18
Sr	660	230	415	574	369	230	230	610
SO ₄	0	<1	14	11	5	16	16	340
HCO ₃	360	250	837	496	2140	-	-	244
Na	30200	15200	33500	29370	41900	37719	37719	43700
K	430	380	554	372	1700	46	46	-
Mg	480	110	374	504	102	873	873	2010
Cl	53000	28000	59100	52360	68000	68083	67959	86900

Table 4: Ions of sea water

Place	Mitchell <i>et al.</i> (1980)	Read and Ringen (1982)	Bezerra <i>et al.</i> (1990)	Todd and Yuan (1992)	McElhiney <i>et al.</i> (2001)		Moghadasi <i>et al.</i> (2002)
	North sea		Campos basin (Brazil)		North sea	Raw S.W	
Ca	403	450	504	428	435	108	267.00
Ba	0	0	1	0	0	0	0.09
Sr	0	9	9	8	0	0	3.40
SO ₄	2480	2300	2834	2960	2860	36	3350.00
HCO ₃	135	170	150	124	-	-	166.00
Na	11000	12100	11500	10890	11424	10665	11750.00
K	340	-	226	460	400	190	-
Mg	1320	1130	1390	1368	1370	475	2996.00
Cl	19800	20950	21300	19700	20635	18170	23000.00

oilfields in the North Sea (Mitchell *et al.*, 1980) that produced barium, calcium and strontium precipitates. They reported that 15-20 μm sized crystals blocked the pore throats in glass bead packs and in alumina cores, by size exclusion and bridging mechanisms. Crystals often grew perpendicular to the pore walls and that some crystal aggregates also had the form of books and rosettes. Todd and Yuan (1992) also conducted laboratory investigations using North Sea reservoir brines that produced barium and strontium scales. Crystals depositing along and growing perpendicular to the pore surfaces caused most of the reduction in core permeability. They observed that doubling the super-saturation ratio of both barium and strontium produced an increase in the quantity of scale formed inside the pores and a change in the morphology of the crystals. Both changes increase the rate of permeability decline.

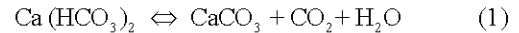
Scale formation along the injection-water path in water-flood operations: At the injection wellhead, injection water temperature is usually much lower than reservoir temperature. When it travels down the injection well-string, the water cools the surrounding formations and its temperature and pressure increase. If the water is saturated at surface conditions with salts whose solubility decreases with increasing temperatures (e.g., anhydrite), scale may form along the well-string.

Scale precipitation from the injection water may happen behind the mixing zone as a consequence of temperature and pressure changes. This is particularly true of waters containing salts whose solubility decreases with increasing temperature and decreasing pressure. Forward of the mixing zone only reservoir brine (with oil) is present in the rock pores. Behind the mixing zone, only injected water in equilibrium at local temperature and pressure (with residual oil) exists. In the mixing zone, precipitation of insoluble salts may occur due to the interaction, at local temperature and pressure, of chemical species contained in the injection water with chemical species present in the reservoir brine.

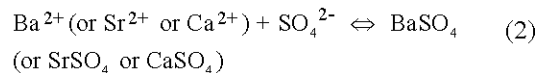
Nevertheless, at a different pressure, the remaining clear water moves ahead mix again with reservoir brine and scale precipitation may again take place. This cycle is repeated until the remaining clear water reaches a production well. Pressure and temperature decrease along the flow string up to the surface in the production well and further changes in thermodynamic conditions occur in the surface equipment. This may again result in scale formation. Normally, these scales do the most damage in the well-bore when there are major falls in pressure but hardly any temperature changes (Khelil *et al.*, 1979).

There are three principal mechanisms by which scales form in both offshore and onshore oil field system (Mackay, 2005; Jordan and Mackay, 2005):

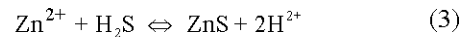
- (a) Decrease in pressure and/or increase in temperature of a brine, goes to a reduction in the solubility of the salt (most commonly these lead to precipitation of carbonate scales, such as CaCO_3).



- (b) 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 BaSO_4).



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



- (c) 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).

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. 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.

Many case histories of oil well scaling by calcium carbonate, calcium sulfate, strontium sulfate and barium sulfate have been reported (Mitchell *et al.*, 1980; Lindlof 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).

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. The chemical species of interest to us are present in aqueous solutions as ions. Certain combinations of these ions lead to compounds, which have low solubility. Once this capacity or solubility is exceeded the compounds precipitate from solution as solids. Therefore, precipitation of solid materials, which may form scale, will occur if:

- The water contains ions, which are capable of forming compounds of limited solubility.
- There is a change in the physical conditions or water composition, lowering the solubility.

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.

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 (Nancollas and Liu, 1975; 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 (Moghadasi *et al.*, 2003a).

MATERIALS AND METHODS

The general purpose of the laboratory test was to investigate permeability reduction by deposition of barium sulfate in a porous medium and knowledge of solubility of barium sulfate and how its solubility are affected by changes in salinity, temperature and pressure.

Core material: The rock cores used in the testes were sandstone from Malaysia with 3 inch length and of diameter 1 inch with average porosity of 32% and of absolute permeability varied from 12.30-13.84 md.

Brines: The ionic compositions of synthetic formation water and water injection (Angsi seawater) are given in Table 5. Note the formation water has barium ions and the sea water contains sulfate ions. It is clear that the mixing of these waters can lead to barium sulfate precipitation.

Scaling test rig: Experiments were carried out using a test rig, which is schematically shown in Fig. 1. The core test equipment consists of five parts: constant pressure pump, transfer cell, oven, pressure transducer and core holder. There follows a brief description of each part.

Constant pressure pump: plunger pump with 1.5 hp motor and the maximum design pressure of the pump is 35 bars was used to injection the brines during flooding at different pressures.

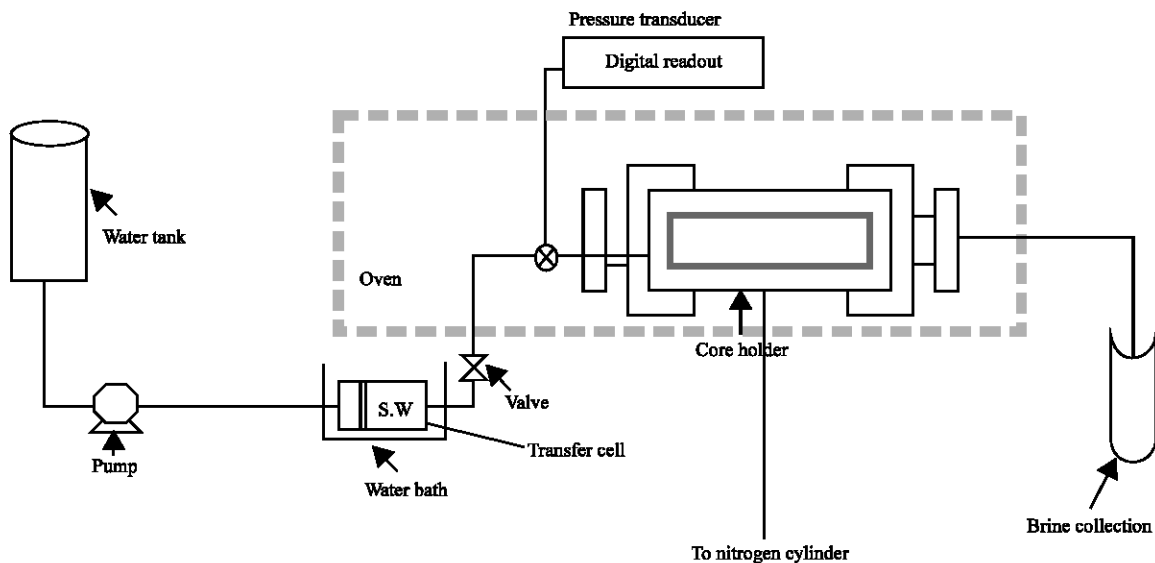


Fig. 1: Schematic of the core flooding apparatus

Table 5: Ions of synthetic formation and injection waters

Ionic	Normal barium formation water (ppm)	High barium formation water (ppm)	Angsi sea water (ppm)
Sodium	41,900	41,900	10,804.500
Potassium	1,700	1,700	375.050
Magnesium	102	102	1,295.250
Calcium	780	780	429.200
Strontium	370	370	6.577
Barium	250	2,200	-
Chloride	68,000	68,000	19,307.450
Sulfate	5	5	2,750.000
Bicarbonate	2,140	2,140	158.800

Transfer cell: Stainless steel transfer cells are used to store and transfer the injection brines to the core holder with a capacity of 1000 mL has a free-floating piston, which separates the pump fluid (distilled water) from the injection brines. The pump fluid is pumped into a transfer cell to displace the brines into the core.

Oven: During all flooding runs, the core holder is placed inside a temperature controlled oven.

Pressure transducer: The differential pressure across the core during flooding runs was measured using pressure transducers (model E-913 033-B29) with a digital display.

Core holder: Stainless steel core holder designed for consolidated core samples 3 inch length and 1 inch diameter was used. A rubber sleeved core holder, subjected to an external confining pressure, into which a sandstone core is placed.

Test procedures

Beaker test: For each experiment of barium sulfate, 100 mL of each filtered opposite waters were poured simultaneously into beaker. The synthetic brines were heated on heat plate and the solution was stirred by magnetic stirrer and after that the solution was filtered through 0.45 µm filter paper. After filtration, 5 mL of the filtrate were pipetted into a 50 mL volumetric flask and filled up to 50 mL. This instantaneous dilution of the BaSO₄ containing brines just after filtration was performed in order to prevent BaSO₄ precipitation during the period between filtering and analytical determination of the Ba concentration. Barium concentrations in the diluted filtrates were determined by atomic absorption spectrometry. The barium determinations were calibrated by measuring five standard solutions was prepared from BaCl₂ solution.

Core test: Before each run, the core sample was heated and dried in preparation for installation in the core-holder.

A vacuum was drawn on the core sample for several hours to remove all air from the core. The core was then saturated with formation water at room temperature and a base permeability determined. After the appearance of formation water at the outlet flooding was continued long enough to ensure 100% saturation. The core holder assembly placed inside the oven and transfer cell containing sea water placed inside the water bath and heated to the desired temperature of the run. The required confining pressure was adjusted to be double inlet pressure. The sea water was injected into the core and mixed with formation water inside porous media. During each run, the flow rate across the core was recorded continuously and the permeability of core was calculated with Darcy’s linear-flow equation before and after scale deposition, in order to observe the effect of scale on well productivity. scale deposition have been observed, the core sample was removed at the end of flooding then dried and cut into sections for Scanning Electron Microscopy (SEM).

RESULTS AND DISCUSSION

Beaker test: Based on the analytical barium concentrations in the diluted filtrates were determined by atomic absorption spectrometry. The solubility of BaSO₄ at various temperatures (40-90°C) and 1 atm of this study were calculated.

The experimental results confirm the general trend in solubility dependencies for BaSO₄ scale with temperatures is obvious and is similar to that observed in the earlier study (Moghadas *et al.*, 2003a). The expected trend in this temperature range is an increase of BaSO₄ solubility with increasing temperature as shown in Fig. 2. The SO₄⁻ content in the sea water brine was reacting with barium ions content in the formation water instantaneously. The more precipitation of BaSO₄ resulting from the presence of a high concentration of barium ions with comparison less precipitation at normal concentration of barium ions.

Core test: The test rig was designed to operate at temperatures of 50 and 80°C. The differential pressure used in these experiments range from 100 to 200 psig.

During each run, the flow rate across the core was recorded continuously and the permeability of core was calculated with Darcy’s linear-flow equation. The flow rate decreased during the experiments only when a super-saturated solution was flowing through the cores. This confirms that the decrease is due to precipitation of the barium sulfate inside the cores with the consequent

reduction in their permeability and porosity. Typical results for flow rate and permeability reduction obtained were plotted versus time and pore volumes of injected brine in Fig. 3-13.

Temperature has a significant influence on solubility and crystal growth of barium sulfate. To study its effect on the permeability reduction, a number of tests were carried out where concentration of injected solution was kept constant at differential pressure from 100 to 200 psig

and temperatures of 50 and 80°C. The Fig. 3 and 8-13 shows the variation of permeability reduction with time and pore volumes of injected brine at different temperatures. Comparing the findings from the scale formation study at 50°C with those obtained from 80°C experiments reveals some aspects of the effects of temperatures. The permeability decline is less rapid at higher temperature, since the rate of precipitation decrease with temperature.

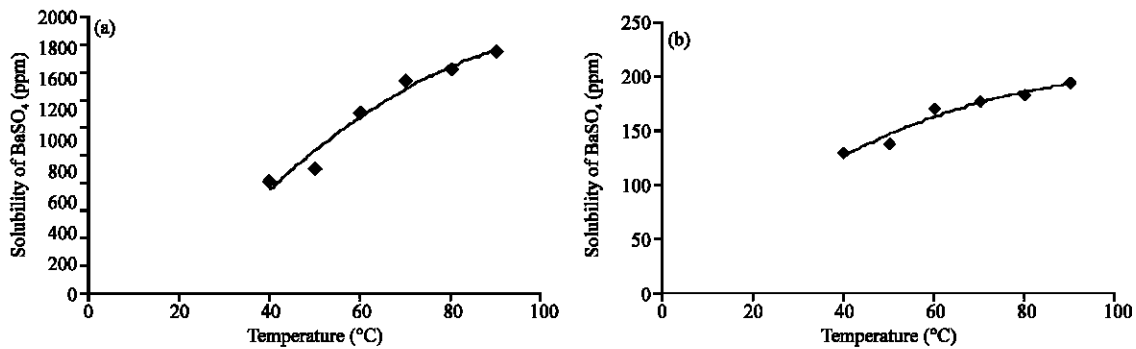


Fig. 2: BaSO₄ solubility is dependent on temperature and (a) Ba = 2200 ppm and (b) Ba = 250 ppm

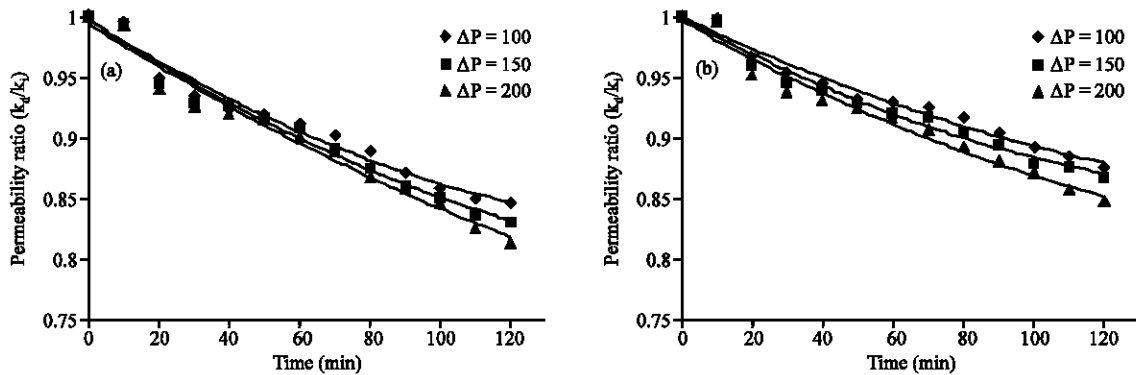


Fig. 3: Permeability ratio vs. time at (a) 50°C and (b) 80°C and Ba = 2200 ppm

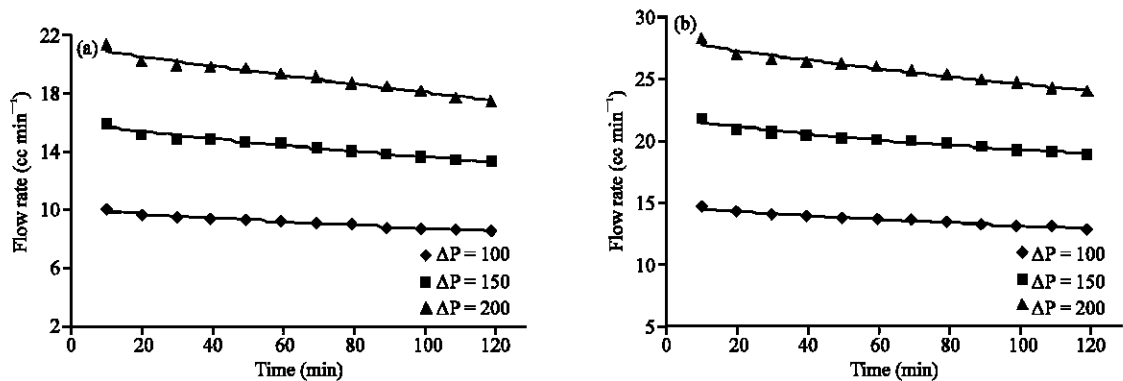


Fig. 4: Flow rate vs. time at (a) 50 and (b) 80°C and Ba = 2200 ppm

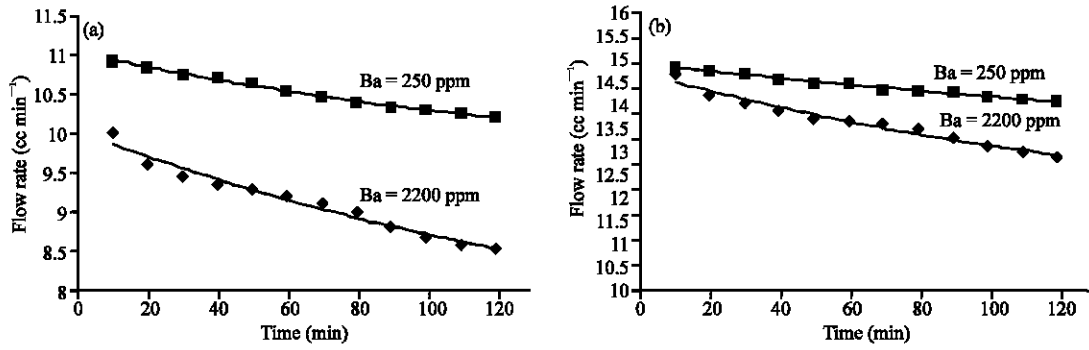


Fig. 5: Flow rate vs. time at (a) 50 and (b) 80°C and $\Delta P = 100$ psig

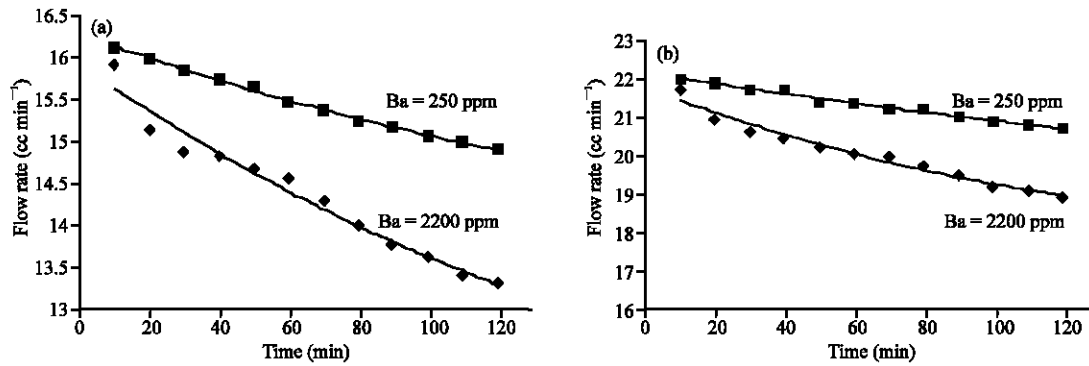


Fig. 6: Flow rate vs. time at (a) 50 and (b) 80°C and $\Delta P = 150$ psig

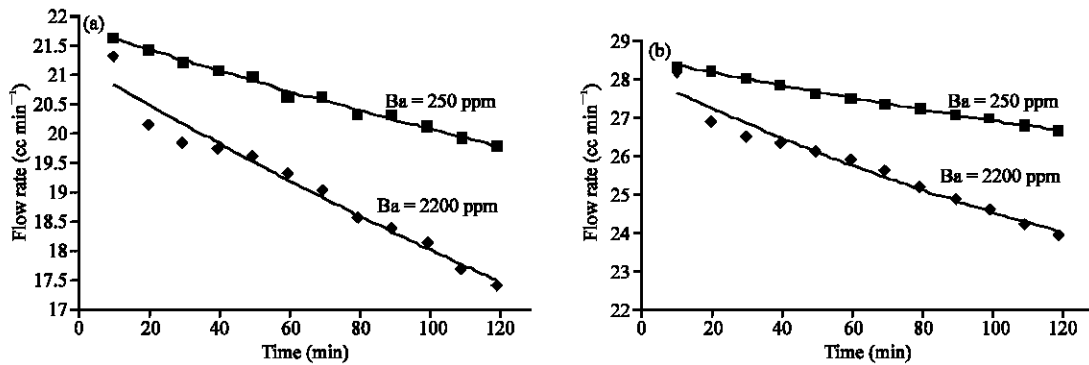


Fig. 7: Flow rate vs. time at (a) 50 and (b) 80°C and $\Delta P = 200$ psig

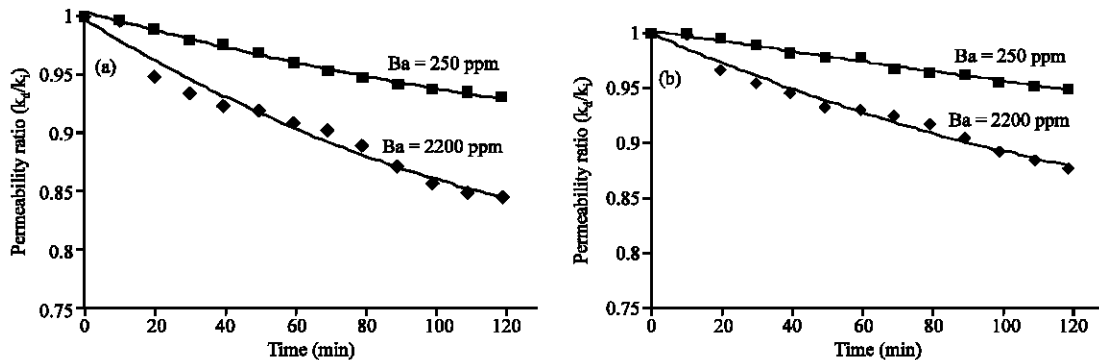


Fig. 8: Permeability ratio vs. time at (a) 50 and (b) 80°C and $\Delta P = 100$ psig

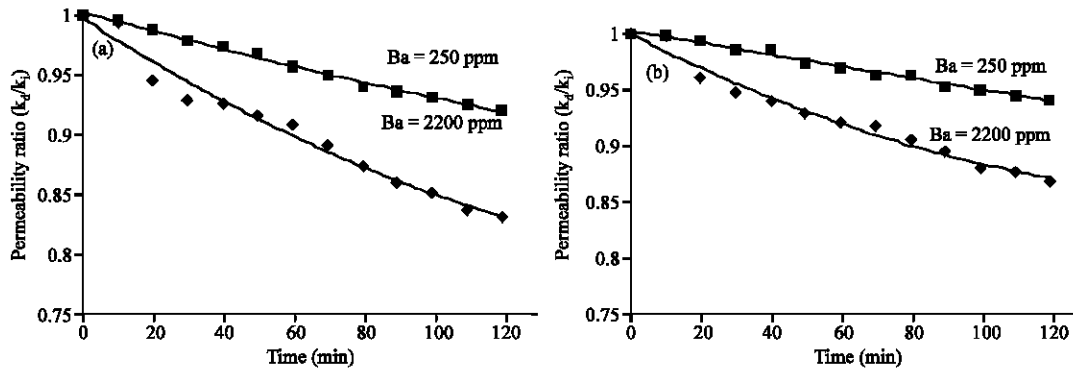


Fig. 9: Permeability ratio vs. time at (a) 50 and (b) 80°C and $\Delta P = 150$ psig

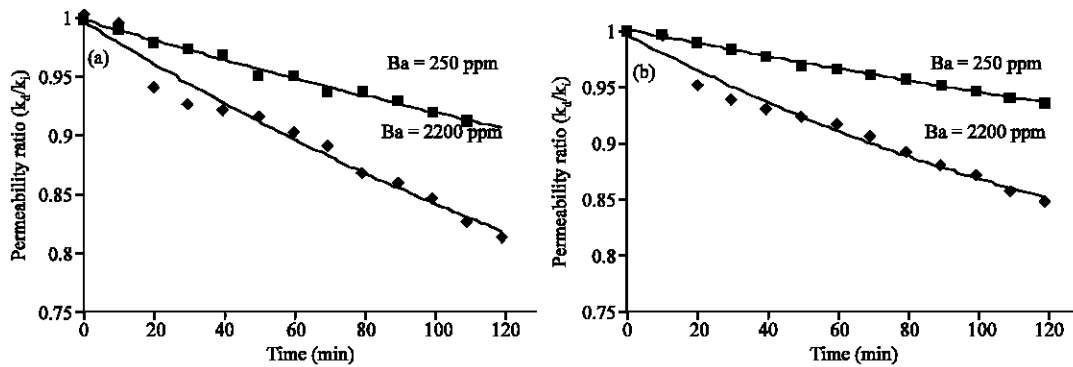


Fig. 10: Permeability ratio vs. time at (a) 50 and (b) 80°C and $\Delta P = 200$ psig

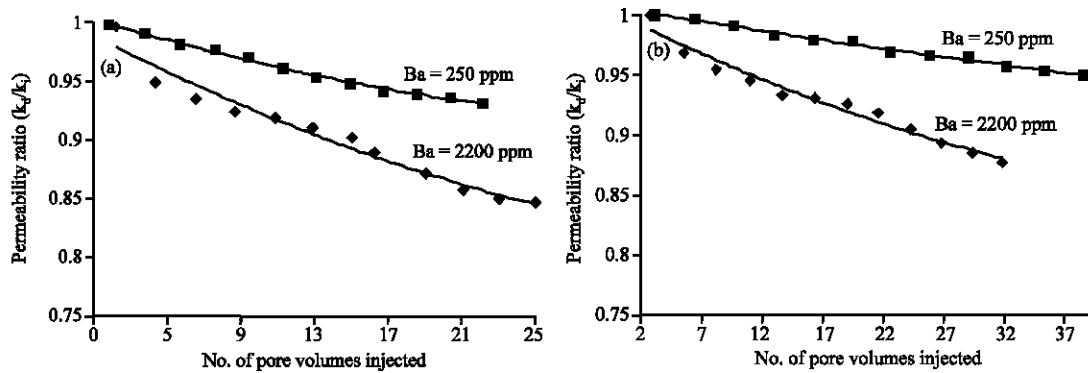


Fig. 11: Permeability ratio vs. pore volumes injected at (a) 50 and (b) 80°C and $\Delta P = 100$ psig

To investigate the effect of flow rate on permeability reduction, depending on the differential pressure used at high differential pressure high flow rate. The permeability decline of porous medium is evident, even at such low flow rates. The overall permeability of porous medium at high barium decreased to between 13 to 19% of initial permeability and normal barium between 5-9% of initial permeability depending on the differential pressure used. At higher flow rates more sulfate ions will enter the porous medium in a given interval of time, hence providing more material for deposition.

A number of tests were carried out to study the effect of solution concentration on permeability reduction. These tests were carried out at differential pressure from 100 to 200 psig and temperatures of 50 and 80°C with two different solution concentrations (high and normal barium, Table 5). Figure 8-13 show the variation in permeability decline with time and pore volumes of injected brine for different concentrations of barium. When the concentration of solution (i.e., super-saturation) is increasing, plugging and hence permeability loss occurs more rapidly. The permeability decline due to high

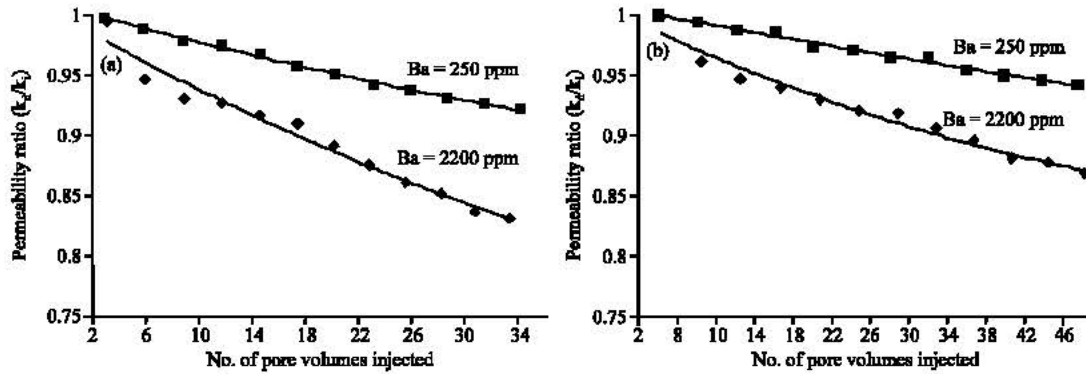


Fig. 12: Permeability ratio vs. pore volumes injected at (a) 50 and (b) 80°C and $\Delta P = 150$ psig

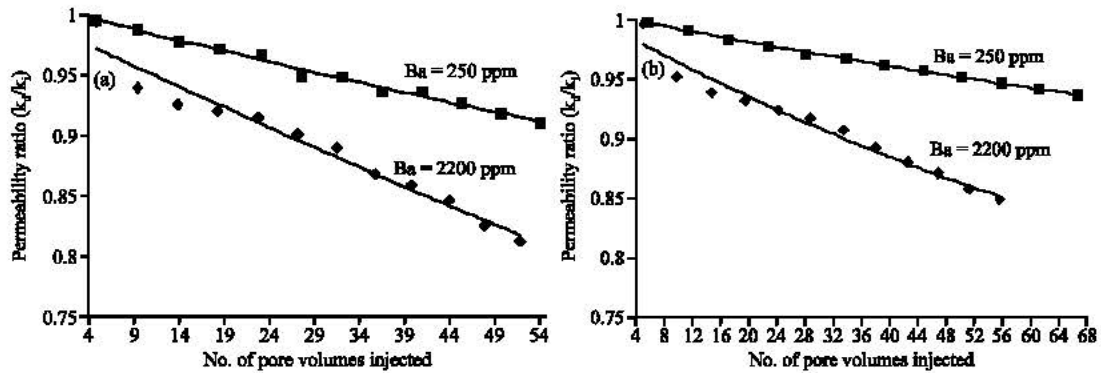


Fig. 13: Permeability ratio vs. pore volumes injected at (a) 50 and (b) 80°C and $\Delta P = 200$ psig

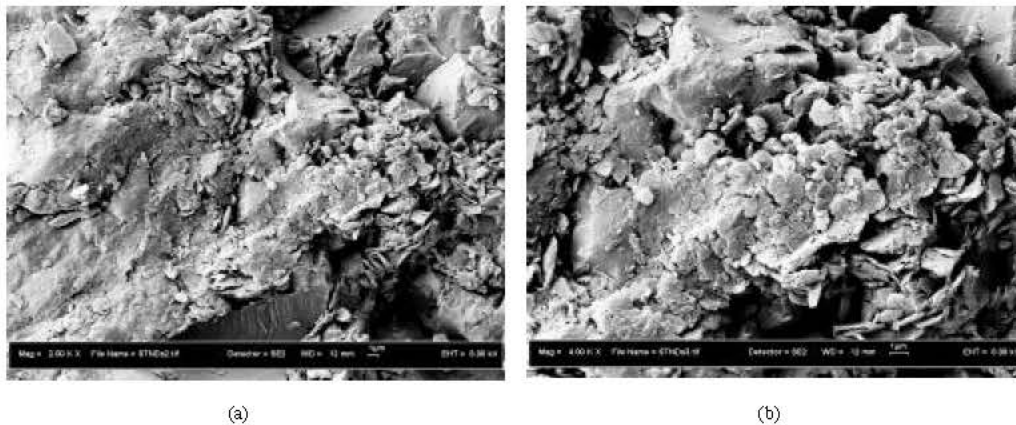


Fig. 14: SEM photograph of an unscaled sandstone core

concentration of barium ions is greater than for normal concentration of barium ions, for given experimental conditions. Figure 14 and 15 shows an SEM photograph of an unscaled core and scaled core samples. SEM photographs revealed the formation of crystals at the inlet face Fig. 15 indicating more precipitation at the inlet face.

Extend of permeability damage: The extend of permeability loss caused by $BaSO_4$ scaling in the rock pores varied in different situations. Figure 8b shows the permeability change of a less damaged core; Fig. 10a shows that of a severely damaged core after $BaSO_4$ scaling. About 5-13% permeability loss is observed in

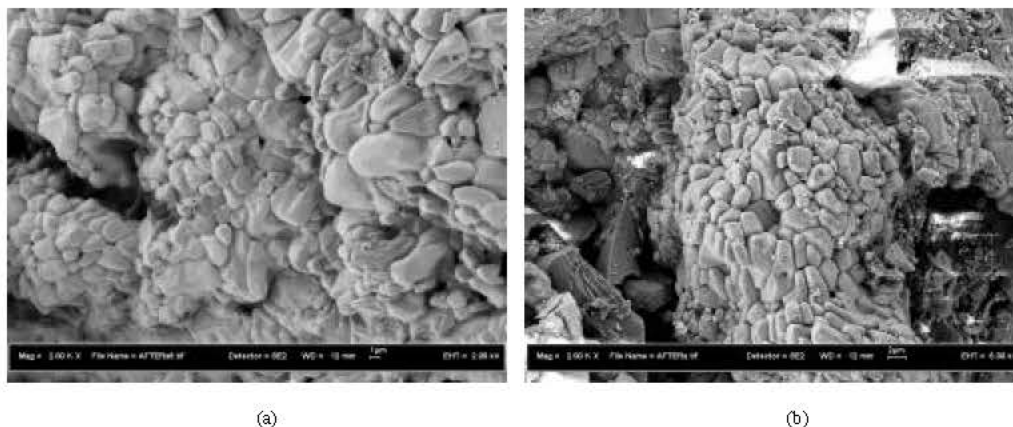


Fig. 15: SEM photograph of BaSO₄ scale in sandstone core

Fig. 8b, but more than 9-19% initial permeability reduction could occur in a heavily scaled core, as Fig. 10a indicates. The reduction in permeability is possibly caused by crystals blocking the pore throats as shown in the SEM view of Fig. 15.

Permeability decline trend: Figure 3 and 8-10 shows the permeability decline trend changes with brine-injection time. During the initial flow period, the permeability declined sharply soon after the two waters mixed in the pores. The permeability decline then slowed and gradually leveled out after the permeability decreased greatly. This phenomenon was observed in all the core tests in which the scaling damage was severe.

CONCLUSIONS

- The experimental results confirm the general trend in solubility dependencies for barium sulfate scales, determined at various temperatures (40 to 90°C) and 1 atm: A temperature rise from 40 to 90°C causes an increase in BaSO₄ solubility
- Permeability decline caused by barium sulfate scale formation in the porous bed ranged from 5 to 19% of the initial permeability, depending on solution composition, initial permeability, temperature and flow rate and solution injection period.
- The pattern of permeability decline in a porous medium due to scaling injection was characterized by a steep initial decline which gradually slowed down to a lower.
- Several factors influencing scale formation had been examined. Increasing temperature, concentration of solution (i.e., super-saturation) and flow rate had a detrimental effect on the permeability reduction.

Pressure had a slight effect on scale formation precipitation.

- The permeability decline due to high concentration of barium ions is greater than for normal concentration of barium ions, for given experimental conditions.

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