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An Experimental Study on Permeability Reduction Resulting from Mixed BaSO₄, CaSO₄, and SrSO₄ Scale Deposition in Porous Media during Water Injection

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Abstract

Sulfate scale deposition (BaSO₄, CaSO₄, and SrSO₄) is a common problem in oilfield operations around the world, which causes significant formation damage during production and injection activities. This paper presents the results of an experimental study on the permeability reduction of porous media due to sulfate scale deposition. A set of experiments were conducted to investigate the effects of cation (Ba²⁺, Ca²⁺, and Sr²⁺ ions) concentration and the number of cation species on the permeability reduction resulting from single sulfate scales (single BaSO₄, CaSO₄, and SrSO₄ scales) and mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition in porous media during water injection. The experiments were performed at a constant temperature of 70 °C and a constant anion (SO₄²⁻ ion) concentration of 3968 ppm in the pack of glass beads as the porous media. The results show that the intensity of permeability reduction increases with increasing cation concentration. These results also declare that the permeability reduction of porous media due to mixed BaSO₄, CaSO₄, and SrSO₄ is clearly severer than single scales.

Keywords: Mixed Scale Formation, Single Scale, Permeability Reduction, Porous Media, Sulfate Scale

1. Introduction

Oil and natural gas demand increases over the time with increasing the global energy consumption. Water injection for maintaining the pressure and improving secondary recovery is a well-established practice in the oil industry. The injected water may impair the permeability or hydraulic conductivity of formation (i.e. formation damage) by fluid-formation or fluid-fluid interactions. A wide range of inorganic scales can be formed by mixing incompatible waters, or due to a change in the temperature, pressure, and pH of a given water stream, which is being injected or produced (Bertero et al., 1988; Bennion, 2002).

Kan and Tomson (2010) have classified common oilfield scales into:

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- pH-independent scales (sulfate scales);
- pH-dependent scales (carbonate and sulfide scales).

pH-dependent scales are acid soluble and their scaling tendency is strongly influenced by the pH of brine. Sulfates (pH-independent scales) may cause considerable permeability reduction, which cannot be solved by acids. Although sulfate scales can be resulted from a change in temperature and/or pressure, when water flows from one location to another, the major cause of sulfate scaling is the chemical incompatibility between sea water (which includes a great amount of sulfate ion) and formation water (the water which exists naturally within the pores of reservoir rock), which originally contains high concentrations of barium, calcium, and/or strontium ions (Yuan and Todd, 1991).

Scale deposition limits oil/gas production by decreasing the area available for fluid flow and plugging the oil-producing formation matrix, fractures, and perforated intervals. Scale also deposits in downhole and production facilities and can cause less well deliverability and more costs for scale removal operations (Bayona, 1993; Moghadasi et al., 2003).

Several experimental studies have been conducted to investigate the permeability reduction of porous media due to single BaSO₄, CaSO₄, and SrSO₄ scales (Essel and Carlberg, 1982; Aliaga et al., 1992, Wat et al., 1992; Moghadasi et al., 2004; Tahmasebi et al., 2007; Jordan et al., 2008; BinMerdhah et al., 2010). A summary of limitations and deficiencies, which have usually existed in the most of these studies are as follows:

- It is difficult and costly to provide core samples for core flooding experiments;
- It is impossible to remove the deposited scales completely from the core plugs without dissolving rock minerals for further experiments;
- A few studies have been devoted to investigate composite scale formation in porous media;
- During core flooding experiments, the trends of permeability reduction at various temperatures and concentrations is usually very close to each other. This is due to low permeability of core sample and the considerable intensity of scale deposition on the injection face of core sample. Regarding this reason, the investigation of the effect of various parameters such as temperature and concentration on the permeability reduction of porous media in core flooding experiments is difficult.

A number of researches were also conducted in the area of composite BaSO₄-SrSO₄ (Read and Ringen, 1982; Bezerra et al., 1990; Todd and Yuan, 1990; Todd and Yuan, 1992), BaSO₄-CaSO₄ (Liu et al., 2012), and CaSO₄-SrSO₄ (BinMerdhah, 2010) scale formation and deposition in dynamic conditions (porous media or small diameter pipes). These studies were performed by means of pack of glass beads/sands, core plugs, and heating coils (with an internal diameter of 1 mm) to investigate the effect of several parameters such as cation concentration and temperature on the rate of permeability reduction of porous media.

Read and Ringen (1982) conducted mixing tests in glass wares and porous media (pack of 4 mm glass beads and synthetic alumina cores) to investigate the effect of strontium and barium mixing ratio on the intensity of scale precipitation. Their research was performed at an NaCl concentration of 40000 mg/l and at temperatures of 20 and 70 °C. Their results showed that the worse permeability reduction occurred for the blend of 90% formation water and 10% sea water. They suggested that deposited scales oriented perpendicular to pore surfaces, which resulted in a greater permeability reduction.

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The co-deposition of BaSO₄ and CaSO₄ in heating coils (dynamic flow apparatus) was studied by Liu et al. (2012). Their experimental research showed that the intensity of composite BaSO₄-CaSO₄ deposition in coil tube was clearly greater than the intensity of single BaSO₄ and single CaSO₄ scale deposition.

Bin Merdhah (2010) examined the $CaSO_4$ and $SrSO_4$ co-precipitation and the effect of some inhibitors on the intensity of scale formation in static tests and core flooding experiments. His study declared that the mass of composite BaSO₄-CaSO₄ precipitation increases with increasing temperature.

Various parameters such as pressure, temperature, flow rate, lithology, and surface properties of porous media and the presence of reservoir hydrocarbon affect the permeability reduction due to scale formation and deposition during water injection. This experimental study was performed to investigate the intensity of scale formation and deposition due to interaction between injected water and formation water without any further interaction such as interaction between hydrocarbon phase and water due to the following reasons:

- According to literature, this paper is the first study on the mixed of three sulfate scale formation in porous media and it was performed to investigate the interaction between the injected water and formation water without any further interaction. Later studies in this area can be performed to investigate the effects of other parameters and compare their effects with the results of the present work.
- Water may be injected in aquifer zone instead of oil zone to provide enough reservoir pressure. Aquifer zone contains formation water without any hydrocarbon.
- The setup described below (section 2.2) provides a low pressure medium for scale formation experiment; therefore, the porous media cannot be saturated with reservoir fluids. Core plugs are needed to study the presence of hydrocarbon phase and they have some limitations as mentioned previously.

Reviewed literature on the scale formation and deposition in porous media showed that the previous studies were devoted to single and the mixture of two scales, while, in this study, the permeability reduction of porous media due to the presence of a mixture of three sulfate scales (BaSO₄, CaSO₄, and SrSO₄) is studied. According to the reviewed literature, this paper is the first study investigating the effects of number of cation species and cation (Ba²⁺, Ca²⁺, and Sr²⁺ ions) concentration on the permeability reduction of a pack of glass beads at a constant temperature of 70 °C. The obtained results were compared with permeability reduction due to single scales to investigate the effect of added cation species on the intensity of permeability reduction. The current paper just deals with an experimental study in the area of mixed scale formation. Later studies can be performed to investigate this phenomenon theoretically. For theoretical discussions in the area of scale formation, the effects of flow rate, concentration, and temperature should be investigated experimentally, while in this paper the effects of the number of cation species and cation concentration have been studied.

2. Materials and methods

2.1. Materials

Brines were prepared by adding predetermined amounts of analytical grade of Na_2SO_4 , $Ca(NO_3)_2.4H_2O$, $Ba(NO_3)_2$, and $Sr(NO_3)_2$ chemicals in micro-filtered distilled water. The chemical composition of solutions used in this study is presented in Table 1.

List of tests.							
Run No.	Solution type	Flow rate (cm ³ /min)	Temperature (°C)	[Ba ²⁺] (ppm)	[Ca ²⁺] (ppm)	[Sr ²⁺] (ppm)	[SO ₄ ²⁻] (ppm)
1	А	50	70	370	0	0	3968
2	В	50	70	740	0	0	3968
3	С	50	70	0	336	0	3968
4	D	50	70	0	0	414	3968
5	E	50	70	370	84	103.5	3968
6	F	50	70	370	168	207	3968
7	G	50	70	740	336	414	3968

Table 1	
ist of tests.	

In this experimental study, sulfate ion concentration was selected based on the chemical analysis of the Persian Gulf water and Mishrif formation water in Siri field (Table 2). The concentrations of cations (Ba²⁺, Ca²⁺, and Sr²⁺) were selected in a way that the effect of cation concentration on the permeability reduction of the pack of glass beads (with a specific size) could be investigated in a proper time. Low calcium ion concentrations (84, 138, and 336 ppm) were selected, since the calcium ion concentrations of Mishrif formation water (>3000 ppm) are so high and cause a severe permeability reduction that these permeability reduction trends will be so close to each other and the effect of cation concentration cannot be distinguished for each cation. Strontium ion concentrations (103.5, 207, and 414 ppm) were also selected based on the reason mentioned about calcium ion concentrations, but they are relatively close to strontium ion concentrations of Mishrif formation water. Barium ion concentrations are higher than the barium ion concentration of Mishrif formation water, but these concentrations are relatively close to the concentration of North Sea. The experiments were performed at a temperature of 70 °C, which is reasonable for Iranian oilfields, and at a flow rate of 50 cm³/min, which is relatively high for Iranian oilfields.

010). Ions (ppm) Siri-C Siri-D Siri-E Sea Water Nosrat Cl-73942 70740 83324 86900 23000 SO_4^{2-} 635 310 142 340 3350 HCO₃-579 528 397 244 166 Mg^{2+} 759 766 5 20 29 Ca^{2+} 4525 8917 7920 3032 267 Na^+ 42215 35391 42800 43700 11750 K^+ 1986 88 -----_____

246

0.09

0.42

3.4

18

610

Water analysis of t	the Persian Gulf wate	er and Mishrif formation	n water in Siri field	(Amiri and Moghadasi, 201
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5.6

760

Table 2

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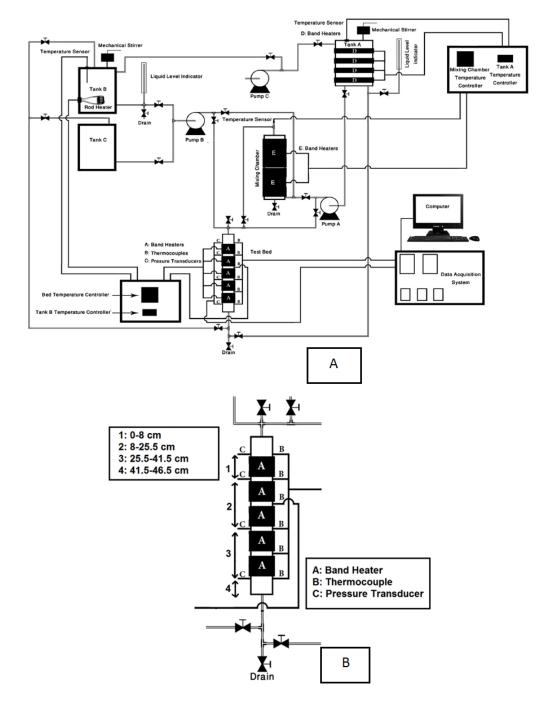
574

Ba²⁺

Fe²⁺

 Sr^{2+}

During the present study, a new version of the test rig, which was introduced by Moghadasi et al. (2004), was designed and re-constructed. The constructed experimental setup is schematically shown in Figure 1. This apparatus consisted of five parts: tanks, pumps, test section (bed), mixing chamber, and data acquisition system.



A schematic representation of: A) Experimental set-up and B) Test section.

The synthetic formation water, which included great amounts of strontium, calcium, and barium ions and the synthetic sea water, which included a great amount of sulfate ions, were stored in separate stainless steel tanks. These tanks were equipped with heaters and digital temperature controllers to adjust an accurate liquid temperature. Two peristaltic pumps were used to inject the distilled water or aqueous solutions into the test section (porous media). The test section was the most important component of this setup, which contained the pack of glass beads as the porous medium. Four pressure transducers and six thermocouples were used to measure the variation of pressure and temperature along the test section. All the transducers and thermocouples were connected to a data acquisition system to monitor and record the experimental data. The flow rate was kept constant during each test and the pressure drop was recorded continuously along the test section. The obtained results were used to calculate the permeability ratio based on Darcy's law. At the end of each experiment, the alkaline solution of ethylene dinitro tetraacetic acid (EDTA) was pumped through the test section to remove the deposited scales and provide the test section for later experiments.

3. Results and discussion

According to Darcy's law, if a linear flow of an incompressible fluid is established through a porous media of length L and a cross-section area of A, then the pressure gradient along the porous media is proportional to the flow rate. This is mathematically defined as (Ahmed, 2006):

$$Q = \frac{K \cdot A}{\mu} \left(-\frac{\Delta P}{\Delta X}\right) \tag{13}$$

Above equation can be rearranged for the permeability of porous media, *K*, as:

$$K = -\left(\frac{Q.\,\mu.\,\Delta X}{A.\,\Delta P}\right) \tag{14}$$

$$K_i = -\left(\frac{Q.\,\mu.\,\Delta X}{A.\,\Delta P}\right)_i\tag{15}$$

where, *K* is permeability at any time and K_i is the initial permeability. During each test, the flow rate (*Q*), fluid viscosity (μ), and physical characteristics of porous media (Δx and *A*) are constant; thus permeability ratio can be defined as:

$$\frac{K}{K_i} = \frac{\left(-\frac{Q.\,\mu.\,\Delta X}{A.\,\Delta P}\right)}{\left(-\frac{Q.\,\mu.\,\Delta X}{A.\,\Delta P}\right)_i} = \frac{\Delta P_i}{\Delta P} \tag{16}$$

For overall section of porous media (0-46.5cm):

A T 7

$$\frac{K}{K_i} = \frac{(P_{out} - P_1)_i}{(P_{out} - P_1)}$$
(17)

Outlet pressure (P_{out}) is equal to the atmospheric pressure (0 psig); hence permeability ratio is defined by the following equation:

$$\frac{K}{K_i} = \frac{P_{1,i}}{P_1} \tag{18}$$

A set of tests were performed to investigate the effect of cation concentration (Ba^{2+} , Sr^{2+} , and Ca^{2+}) and the number of cation species on the permeability reduction of porous media due to scale deposition. These tests were conducted at a temperature of 70 °C, which was reasonable for Iranian oilfields, and at a flow rate of 50 cm³/min. During each experiment, the pressure drop along the test section was recorded continuously and then the permeability ratio was calculated by using Equation 18.

3.1. Barium sulfate scale formation

A set of tests were performed to investigate the effect of barium ion concentration on the permeability reduction of porous media due to single $BaSO_4$ scale formation and deposition. Figure 2 shows the variation of permeability reduction as a function of the pore volume injected of brine at a constant temperature of 70 °C and at barium concentrations of 370 and 740 ppm. The results declare that at a higher barium ion concentration, the rate of permeability reduction increases.

Figure 2 shows that an almost 25% permeability reduction was occurred for the lower barium ion concentration test (Run#1), while a 47% permeability reduction was observed by increasing barium ion concentration (Run#2). As the barium ion concentration increases, the intensity of permeability reduction rises due to:

- 1- The supersaturation of the brine increases, when anionic and cationic solutions are mixed in porous media; consequently, the rate of nucleation and scale precipitation increases.
- 2- The rate of crystal growth increases and thus larger scales are deposited in porous media and a greater permeability reduction is observed.

These results are in good agreement with previous studies (Read and Ringen, 1982; Todd and Yuan, 1990; BinMerdha et al., 2010).

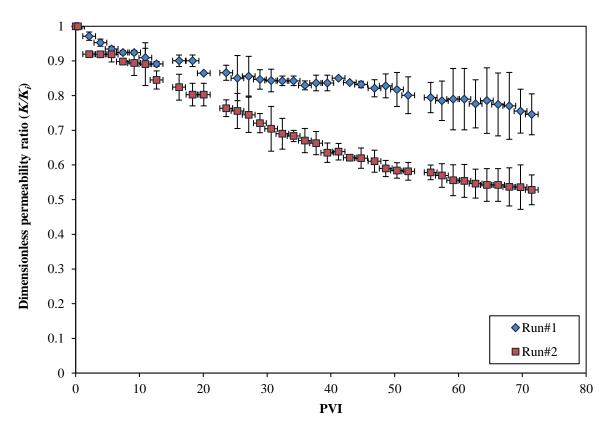


Figure 2

Variation of permeability ratio and the standard error of each run as a function of the pore volume injection of brine showing the effect of barium ion concentration.

On the other hand, the permeability decline trends in Figure 2 demonstrate that the permeability reduction rate (the slope of permeability reduction trends) at the first stage of the experiments is

relatively high, but these rate decreases by increasing the pore volume injected of brine (time of injection). This phenomenon declares that the single $BaSO_4$ deposits are very weak and they are detached from their places in the pack of glass beads by increasing the pressure drop along the test section; consequently, the severity of scale deposition and permeability reduction decrease after a certain period of injection.

To describe the repeatability of the reported data and the amount of the uncertainty of the measurements, the standard errors (the standard deviation from the mean) of each scale formation test have been added to Figures 2 and 3 in the form of error bars. The error bars in Figure 2 show that the magnitude of the standard error rises by increasing the pore volume injected of brine, because at the first stage of the scale formation experiments, the permeability and the rate of permeability reduction are high; hence the difference between the scale deposition and permeability reduction in the conducted tests is not clear, while, as the permeability of the porous media drops by increasing the pore volume injected of brine, a small difference in scale formation and deposition between the conducted tests can make a greater difference in the amount of their permeability reduction. Consequently, the magnitude of standard error rises by increasing the pore volume injected of brine. According to this figure, the standard errors of the repeated tests are relatively reasonable.

3.2. Calcium sulfate scale formation

Since the calcium ion concentrations selected for these experiments were too low, the results did not show a measurable reduction in permeability.

3.3. Strontium sulfate scale formation

According to the literature (Jacques et al.,1983; Lindlof and Stoffer, 1983), strontium sulfate scale formation rises by increasing strontium ion concentration, but no SrSO₄ precipitation was even observed during the experiments at the highest strontium ion concentration selected for this study (i.e. $[Sr^{2+}] = 414$ ppm).

3.4. Mixed BaSO₄, CaSO₄, and SrSO₄ scale formation

In this section, the effect of cation $(Ba^{2+}, Sr^{2+}, and Ca^{2+})$ concentration on the permeability reduction of porous media due to mixed BaSO₄, CaSO₄, and SrSO₄ precipitation is investigated. A number of tests (Runs 5-7 presented in Table 1) were conducted at a temperature of 70 °C and various concentrations of barium, strontium, and calcium ions. Figure 3 shows that the rate of mixed scale precipitation and consequently permeability reduction increases as cation concentration rises.

As it was mentioned in section 3.1, the intensity of permeability reduction rises by increasing cation concentration, since the supersaturation of brine and crystal growth increase as the cation concentration rises. An almost 68-85% permeability reduction is observed in Figure 3 during the mixed scale formation experiments, which is so high. Figure 3 also demonstrates that the reduction rate of the permeability loss (the slope of permeability decline trends) is clearly lower than the single BaSO₄ scale formation experiments. In other words, the rate of permeability reduction is almost constant during water injection. The results show that the trends of permeability reduction in the mixed scale formation experiments, the permeability decline trends at various barium ion concentrations are clearly separated and distinguished. This is due to the fact that the high intensity of permeability loss in the mixed cases does not let the permeability decline trends to show their

distinction from each other. However, the rate of the mixed scale deposition and consequently formation damage increases by raising cation concentration.

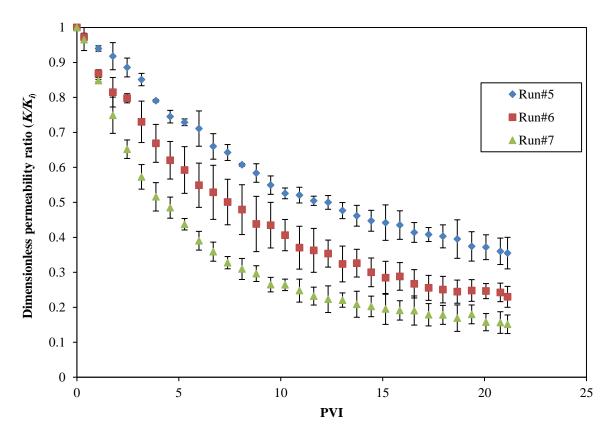
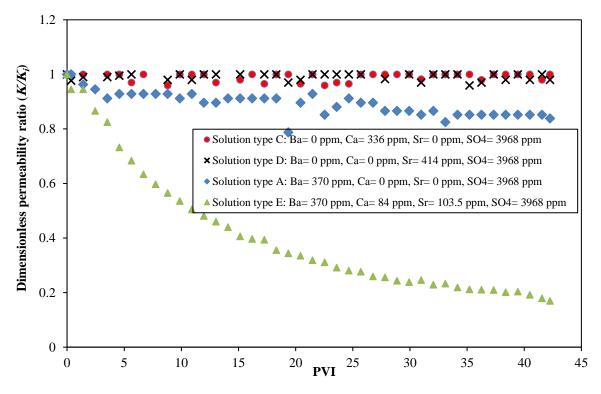


Figure 3

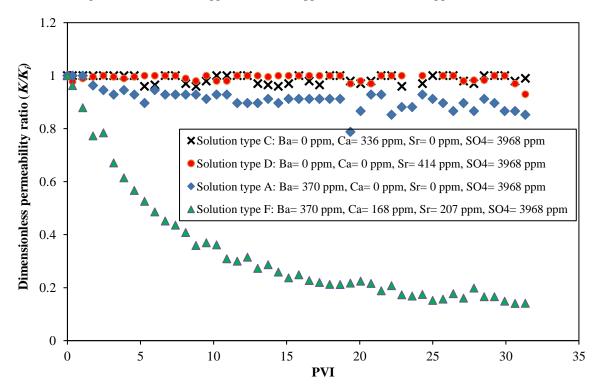
Variation of permeability ratio and the standard error of each run as a function of the pore volume injection of brine showing the effect of cation concentration.

As Figure 3 shows, in the case of the mixed scale formation experiments, the standard errors also grow by increasing the pore volume injected of brine, but in this case the intensity of the scale formation and deposition is clearly greater than the single scale formation experiments; thus the permeability drops rapidly and the magnitude of the standard error is also high at the first stage of the injection. According to this figure, the relative errors of the repeat tests are relatively reasonable.

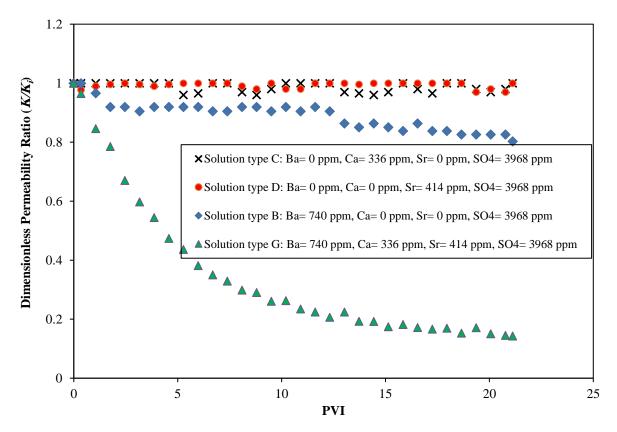
Permeability reduction resulting from the mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition is compared with the permeability reduction due to single scales (single BaSO₄, CaSO₄, and SrSO₄ scales) in Figures 4-6. In these figures, the maximum concentrations of strontium and calcium ions selected for this study ($[Sr^{2+}] = 414$ ppm and $[Ca^{2+}] = 336$ ppm) were considered for single CaSO₄ and SrSO₄ scale deposition experiments. The permeability reduction of porous media caused by these scales at lower cation concentrations was also negligible. It can be observed that the intensity of permeability reduction increases by adding cation species. In other words, at similar operational conditions, the rate of permeability reduction of porous media, due to mixed scales, is greater than single scales.



Variation of permeability ratio as a function of the pore volume injection of brine showing the effect of the number of cation species at $[Ba^{2+}] = 370$ ppm, $[Ca^{2+}] = 84$ ppm, and $[Sr^{2+}] = 103.5$ ppm.

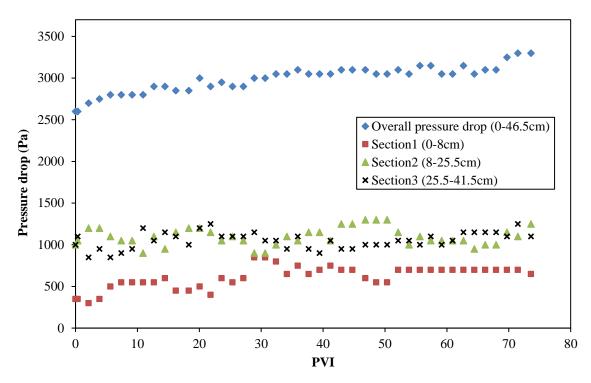


Variation of permeability ratio as a function of the pore volume injection of brine showing the effect of the number of cation species at $[Ba^{2+}] = 370$ ppm, $[Ca^{2+}] = 168$ ppm, and $[Sr^{2+}] = 207$ ppm.

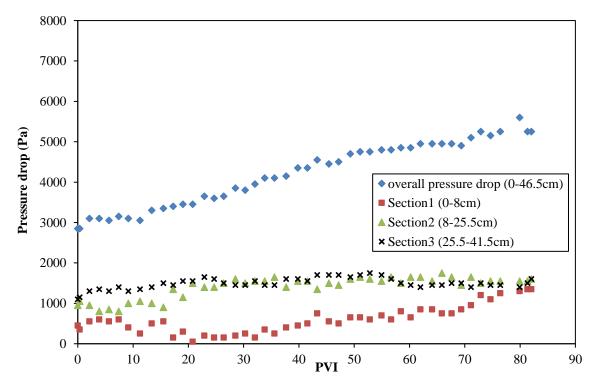


Variation of permeability ratio as a function of the pore volume injection of brine showing the effect of the number of cation species at $[Ba^{2+}] = 740$ ppm, $[Ca^{2+}] = 336$ ppm, and $[Sr^{2+}] = 414$ ppm.

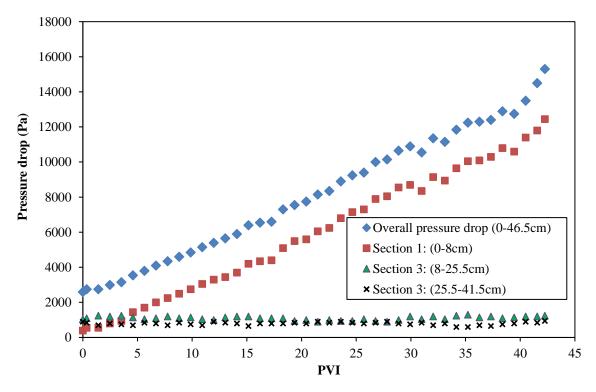
Figures 7-11 show the variation of pressure drop along the porous media as a function of the pore volume injected of brine. Pressure drop trends declare that the single barium sulfate deposition occurs almost in all the sections of the porous media (Figures 7 and 8), while the mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition observed only in section 1 (0-8 cm) of the test section (Figure 9-11). For better understanding of this issue, the contribution of each section of the porous media to an increasing overall pressure drop along the test section during the scale formation experiments is presented in Figure 12 (A-E). The parts A and B of this figure show that all the sections of the porous media have experienced an increase in pressure drop during single BaSO₄ scale formation experiments. For example, 43%, 36%, and 14% increases in overall pressure drop are respectively due to the contributions of sections 1, 2, and 3 for Run#1. The parts C-E of Figure 12 demonstrate that an increase larger than 95% in overall pressure drop is due to scale deposition in the section 1 of the test section, while the role of the other sections is negligible. These results confirmed that the mixed crystals were different in physical properties from barium sulfate deposits; in other words, mixed scales are larger than single scales.



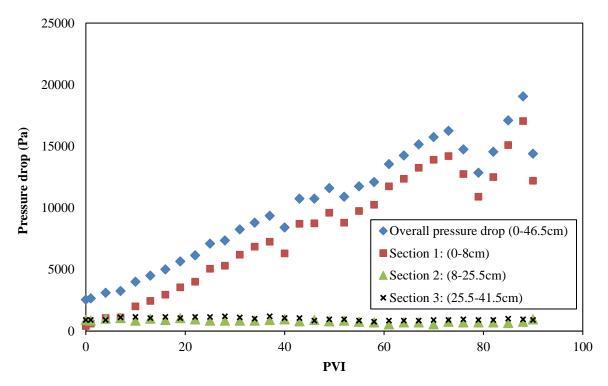
Variation of pressure drop as a function of the pore volume injection of brine due to single $BaSO_4$ scale deposition at $[Ba^{2+}] = 370$ ppm.



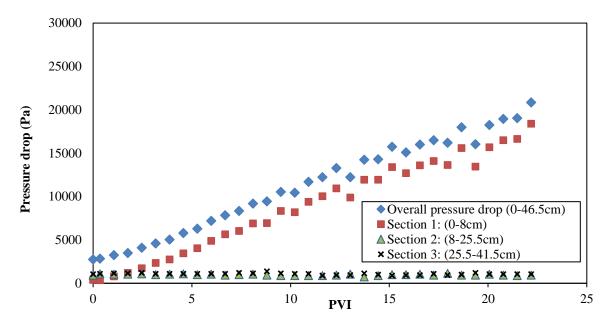
Variation of pressure drop as a function of the pore volume injection of brine due to single $BaSO_4$ scale deposition at $[Ba^{2+}] = 740$ ppm.



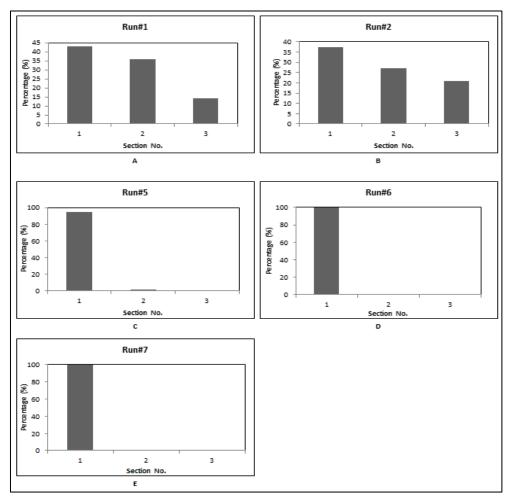
Variation of pressure drop as a function of the pore volume injection of brine due to mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition at $[Ba^{2+}]=370$ ppm, $[Ca^{2+}]=84$ and $[Sr^{2+}]=103.5$ ppm.



Variation of pressure drop as a function of the pore volume injection of brine due to mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition at $[Ba^{2+}]=370$ ppm, $[Ca^{2+}]=168$ and $[Sr^{2+}]=207$ ppm.



Variation of pressure drop as a function of the pore volume injection of brine due to mixed BaSO₄, CaSO₄, and SrSO₄ scale deposition at $[Ba^{2+}] = 740$ ppm, $[Ca^{2+}] = 336$ and $[Sr^{2+}] = 414$ ppm.



Contribution of each section of the porous media to increasing overall pressure drop along the test section.

It should be noted that the amount of the pore volume injected of brine (durations of injection time) in various cases (single and mixed scale formation experiments) are herein different to demonstrate that the rate of permeability reduction due to single sulfate scale formation is clearly slower than mixed scales. In other words, in the case of the single scale formation experiments, it takes time to reach a measurable and proper permeability level to be compared with the other cases, while in the mixed scale tests, the permeability reduction occurs fast. However, the amount of the pore volume injection of brine (durations of time) in Figures 4-6 were equalized to compare the intensity of formation damage due to single and mixed sulfate scale formation.

When anionic and cationic solutions were injected through the porous media, $BaSO_4$ scales were quickly formed and they acted as nucleation locations for $CaSO_4$ and $SrSO_4$ deposition. At higher cation concentrations, the influence of $CaSO_4$ and $SrSO_4$ precipitation on the $BaSO_4$ crystals increased, and thus the permeability reduction of the porous media occurred more rapidly.

The reaction of alkaline solution of EDTA with the deposited scales suggested that mixed BaSO₄, CaSO₄, and SrSO₄ scales were clearly different from single scales in chemical properties. The observations declared that a white color solution was formed due to the reaction of EDTA solution with the barium sulfate deposits, while the reaction of the injected EDTA solution with the mixed BaSO₄, CaSO₄, and SrSO₄ scales at a high cation concentration resulted in the formation of a colorless solution. At a low cation concentration, the impact of CaSO₄ and SrSO₄ deposition on the BaSO₄ crystals decreased, and thus the chemical properties of the mixed scales were nearly similar to those of single BaSO₄ scales and a ghost-white color solution was formed due to the reaction of EDTA solution with the mixed scales.

4. Conclusions

- The effect of cation concentration on the mixed scale formation was studied and the results declared that even minute amounts of strontium and calcium ions combined with barium ions could make a considerable permeability reduction, while permeability reduction caused in single scale cases (single CaSO₄ or SrSO₄ scale precipitation experiments) was negligible.
- A higher scaling rate, deposition, plugging, and therefore permeability reduction were observed by adding cation species.
- Pressure drop as a function of the pore volume injected of brine along the test section showed that BaSO₄ precipitation occurred almost in all the sections of the porous media (test section), while the mixed scale deposition was observed only in the section 1 (0-8 cm) of the test section. These results confirmed that the mixed scale crystals were larger than single BaSO₄ precipitates.

Nomenclature

Α	: Cross-sectional area (m ²)
EDTA	: Ethylene dinitro tetraacetic acid
Κ	: Current permeability (m ²)
K_i	: Initial permeability (m ²)
Р	: Pressure (N/m ²)
PVI	: Pore volume injected
Q	: Volumetric flow rate (m ³ /s)

X	: Distance (m)
Greek	
μ	: Viscosity (kg/m.s)

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