

# EXPERIMENTAL AND MODELING INVESTIGATION OF ION EXCHANGE DURING LOW SALINITY WATERFLOODING

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## ABSTRACT

In recent years, the salinity and composition of the invading and connate brine has been found to be critical for oil recovery. Low salinity water has been reported as being capable of improving oil recovery in sandstone cores under certain conditions. Though the exact mechanism is still not clearly understood, a number of authors have suggested various mechanisms. A common feature in the suggested mechanism is the release of divalent cations from the rock surface. Examples of proposed mechanisms where this release of divalent cations can be important include:

- 1.) Removal of oil components bounded to these ions as described by multicomponent Ion Exchange mechanism.
- 2.) Increased local pH.
- 3.) Clay dispersion.

In this paper, we present one phase laboratory experiments of waterfloods in Berea sandstone cores using brines with different compositions and static experiments with reservoir rock and brines of different compositions. In each of these experiments, it was observed that low salinity core flood leads to a release of cations from the core as against flooding with more saline brines.

A model is subsequently described that predicts the release of ions in low salinity water floods and relates this to improved displacement and recovery. More precisely, the model assumes the released cations to be principally from the clay minerals found in sandstone rocks. The amount released is calculated based on a fast ion exchange equilibrium calculation that is salinity dependent. The amount of divalent ions released by brine-rock interaction as predicted by the model is found to correlate with low salinity increased recovery from literature. This model can be used for screening optimal ion concentrations for low salinity floods.

## INTRODUCTION

In the past decade a lot of interest has been generated on the effect of brine salinity on oil recovery. Several authors have published results in which low salinity as invading brine improved recovery as against invading with more saline brine in sandstone rock. The exact mechanism in which low salinity brine improves the recovery is still not widely accepted though a number of mechanisms have been suggested. Among these

mechanisms are multicomponent ion exchange mechanism [1], increased local pH as described by Austad [2] and clay dispersions [3].

A common feature of these mechanisms is the release of divalent ions from negatively charged sites (very often clays). This paper presents results showing the release of divalent cations in a one phase waterflood and also describes a mechanism which couples the flow behavior in a two-phase flow to desorption of divalent ions after a fast salinity-dependent ion exchange.

## LITERATURE REVIEW

Traditionally the chemistry of injected water in sandstone reservoirs was thought to be of little or no importance except if these reservoirs contain swelling clays. Yildz and Morrow [4] showed that the brine composition had an effect on oil recovery. Various authors [3,5] have shown that a decrease in connate water salinity significantly improved the oil recovery after waterflood. Since then a number of authors have documented improved recovery after waterflood by reducing the salinity of the invading brine [6, 7]. The following have been listed as the requirement of low salinity improved recovery:

- ✓ Presence of clay [3]. However, Pu [8] showed low salinity improved recovery in cores without significant clay content but with dolomite. This suggests that at least some negative charged surfaces are needed.
- ✓ Use of crude oil containing some polar component [3]. Austad [2] showed the low salinity effect on both an acidic based crude oil and a basic based crude oil
- ✓ Presence of connate brine [3]. Lager [1] and Sharma [5] did not find the low salinity effect in secondary core floods if the connate water contained no divalent ion.

However there have been core-flood experiments carried out meeting the above conditions and yet no appreciable improved recovery was observed [9].

Several mechanisms have been proposed in the literature to explain the low salinity effect. Among these mechanisms are:

1. Multicomponent Ion exchange mechanism [1]: This mechanism stipulates that polar component in the oil phase can be adsorbed to the clay surface by ion binding with the divalent ion in the water phase. Low salinity water is proposed to act in such a way as to expand the double layer and make the divalent ions and its associated oil component ready for exchange by incoming ions.
2. Dispersion of clay as a result of expansion of the double layer. It is thought that the dispersed clay results in the release of the oil components that were initially attached to the clay. See Tang and Morrow [3].
3. Dissolution of oil wet soluble mineral surfaces, especially minerals acting as cements in the clays. Such dissolution is stronger at lower ionic strength [10].
4. pH Increase: Morrow [3] reported that low salinity induced recovery is usually accompanied by a pH increase to about 8 and suggested that the mechanism of this to be a combination of ion exchange and carbonate dissolution. McGuire [11] further links generation of surfactants, changes in interfacial tension and wettability changes obtained in the high pH environment to be responsible for the improved recovery. Austad [2] proposed a chemical mechanism in which desorption of divalent cation ultimately leads to a 'local' increase in pH and subsequent desorption of adsorbed

polar oil component. However Lager [1] states that increased pH induced recovery can not be the mechanism of low salinity because increased pH has not been observed in all low salinity experiments and the difficulty in replicating the pH seen in corefloods to reservoir conditions where pH buffering would most likely occur.

Buckley [10] has stated that the low salinity effect is more pronounced with low salinity connate water and hence any mechanism that is based solely on changing from high salinity to low salinity brine (Osmotic pressure mechanism, salinity shock and multi ion exchange mechanism) must be discounted.

**Divalent Ion and Wettability:** There are several mechanisms in which organic matter can be adsorbed on mineral surfaces. These mechanisms include: van der Waals interaction, ligand exchange, cation bridges, columbic interaction (anion and cation exchanges) and hydrophobic/hydrophilic effect [12, 13]. The importance of each of these mechanisms can vary depending on crude-oil-brine interaction. In a study of pore water – organic matter-montmorillonite interaction, Arnarson [13] estimated the adsorption to be due to 60% van der Waals forces, 35% ligand exchange and 5% cation bridges. However Tipping [14] found that divalent cation presence can improve the adsorption of organic substances on iron oxides by about 100%. The divalent cations can play a significant role in the cation exchange, cation bridging and ligand exchange mechanisms of adsorption [14, 15]. It has been reported that low salinity effect is not seen if cores were aged without divalent cation [1, 16].

## DESCRIPTION OF EXPERIMENT

Two kinds of experiments were carried out to investigate ion exchange: static experiments and dynamic/flooding experiments.

**Dynamic Experiment:** Brines were prepared by dissolving the appropriate mass of salt in distilled water. The compositions of the brines used in all the experiments are given in Table 1. Berea core of length 30cm, diameter 1.5 inch, porosity 0.23 and permeability 500mD was used. The experiment was set up as shown in Figure 1. A succession of formation water, sea water and low salinity water was flooded through the core at 80°C using an injection rate of 0.05ml/min. Effluent samples were collected at intervals and analyzed for elements using Inductive Coupled Plasma (ICP).

**Static Experiment:** The interactions of reservoir rock were studied in static experiments by mixing the crushed minerals and brines in a test-tube then stored in the oven at 80°C. The samples were mixed at intervals. Each sample was prepared with 2g of the mineral and 5g of brine. Several samples of reservoir rock and each of the brines were stored in the oven. Minimum 2.0 ml sample of brine was taken from the first tube after 1 day and from the remaining test-tubes after 7 days and 14 days respectively. The samples taken out were analyzed for selected elements using ICP. The experiment was repeated at 40°C to study the effect of temperature.

## RESULTS

The result from the dynamic experiment is shown in Figure 2a. The result shows that the potassium concentration during formation water and sea water injection is relatively unchanged from the injected value. There is also a release of magnesium ion during the

early stage of formation water injection which maybe due to an ion-exchange. There is some release of calcium ion throughout the duration of formation water injection. This suggests some carbonate dissolution. The seawater injection did not show any appreciable release of ions. During the low salinity water injection (Figure 2b) there is a rather slow return to the injected concentrations of the ions. This point to the release of ions from the core.

The static result shows (Figure 3) that there is much more release of calcium ion in low salinity water and distilled water than there is release of potassium. Though the reason for the release is not clear, it is most likely due to desorption and dissolution.

### Ion Exchange-Transport Model

Here we develop a model that calculates the amount of ions that are attached to the rock surface during transport with particular interest in the divalent ions. The aim is to link subsequent desorption of the divalent ions to improved flow functions as suggested by multicomponent ion exchange mechanism [1] and chemical mechanism by Austad [2]. A one dimensional flow of the phases and ions is given by:

$$\partial_t(\phi s_o) + \partial_x(-k\lambda_o[(p_o)_x - \rho_o g]) = 0 \quad (\text{Oil Transport}) \quad (1)$$

$$\partial_t(\phi s) + \partial_x(-k\lambda[(p_w)_x - \rho g]) = 0 \quad (\text{Water Transport}) \quad (2)$$

$$\partial_t(\phi s C_{na}) + \partial_t(M_c \beta_{na}) + \partial_x(-k C_{na} \lambda[(p_w)_x - \rho g]) = \partial_x(D(\phi, s) \partial_x C_{na}) \quad (3)$$

$$\partial_t(\phi s C_{mg}) + \partial_t(M_c \beta_{mg}) + \partial_x(-k C_{mg} \lambda[(p_w)_x - \rho g]) = \partial_x(D(\phi, s) \partial_x C_{mg}) \quad (4)$$

$$\partial_t(\phi s C_{ca}) + \partial_t(M_c \beta_{ca}) + \partial_x(-k C_{ca} \lambda[(p_w)_x - \rho g]) = \partial_x(D(\phi, s) \partial_x C_{ca}) \quad (\text{Ions in Water}) \quad (5)$$

$$\partial_t(\phi s C_{so}) + \partial_x(-k C_{so} \lambda[(p_w)_x - \rho g]) = \partial_x(D(\phi, s) \partial_x C_{so}) \quad (6)$$

$$\partial_t(\phi s C_{cl}) + \partial_x(-k C_{cl} \lambda[(p_w)_x - \rho g]) = \partial_x(D(\phi, s) \partial_x C_{cl}) \quad (7)$$

Where  $C_{na}$ ,  $C_{mg}$ ,  $C_{ca}$ ,  $C_{cl}$ ,  $C_{so}$  are the concentrations of sodium, magnesium, calcium, chloride and sulphate ions respectively.  $\beta_{na}$ ,  $\beta_{mg}$ ,  $\beta_{ca}$ , are the sodium, magnesium and calcium ions that are adsorbed by the clay. These are determined using the Gapon ion-exchange model [17] given by:



A mass action equation can be written for the exchanges as

$$K_{cana} = \frac{\beta_{ca} \gamma_{na} C_{na}}{\beta_{na} \sqrt{\gamma_{ca} C_{ca}}} \quad (10)$$

$$K_{mgna} = \frac{\beta_{mg} \gamma_{na} C_{na}}{\beta_{na} \sqrt{\gamma_{mg} C_{mg}}} \quad (11)$$

where  $K_{mgna}$  and  $K_{cana}$  are the selectivity factors. Hirasiki[18] describes a model that describes the selectivity factors as a function of the electrolyte. In this case we simply

treat the selectivity factors as a function of brine salinity. The sum of the ions adsorbed on the clay is at all times equal to the cation exchange capacity

$$\beta_{na} + 2\beta_{mg} + 2\beta_{ca} = CEC \quad (12)$$

In addition to the above equations (13) and (14) deals with the capillary pressure and saturation respectively.

$$P_c = p_o - p_w \quad (13)$$

$$s_o + s_w = 1 \quad (14)$$

With equations (1)-(7) and (10)-(14) we have 12 equations to solve 12 unknowns ( $s_o$ ,  $s_w$ ,  $p_o$ ,  $p_w$ ,  $C_{na}$ ,  $\beta_{na}$ ,  $C_{mg}$ ,  $\beta_{mg}$ ,  $C_{ca}$ ,  $\beta_{ca}$ ,  $C_{so}$ , and  $C_{cl}$ ).

$\lambda_o$ ,  $\lambda$  and  $P_c$  are the flow functions which ordinarily are saturation dependent. Tripathi et al. [19] modeled these functions to be salinity dependent in addition to saturation. Yu et al. [20] modeled it as function of a wettability alteration agent. For reasons earlier described, we chose to model it as dependent on desorption of the divalent ion in addition to saturation.

A weighting function  $F$  is defined by the desorption of divalent ions as such

$$F(\beta_{ca}, \beta_{mg}) = \frac{1}{1 + Km(\beta_{ca}, \beta_{mg})} \quad (15)$$

where  $K$  is a constant and  $m$  is the sum desorption of divalent ion given by

$$m(\beta_{ca}, \beta_{mg}) = \max[(\beta_{cao} - \beta_{ca}), 0] + \max[(\beta_{mgo} - \beta_{mg}), 0] \quad (16)$$

Where  $\beta_{cao}$  and  $\beta_{mgo}$  are the original calcium and magnesium adsorbed on the rock. Equation (15) is such that  $F = 1$ , when there is no desorption of calcium and magnesium ions from the rock and  $0 < F < 1$ , when there is some desorption of any of calcium or magnesium ions. The weighting function,  $F$  is tied to different flow functions as illustrated in Figure 4 similar to [19, 20]. Full details of the model will be published soon. The extreme of the flow functions (when  $F = 0$ ) corresponding to maximum desorption of the divalent may not be readily available except it is determined experimentally. The model is very useful in identifying different brine composition and rock properties that will lead to desorption of divalent ion and hence improved flow functions and oil recovery. Examples are presented below.

**Example One:** No divalent Ions in Connate/ageing Brine

Lager et al. [1] and Ligthelm et al. [16] performed corefloods in which the connate brine had no divalent ion. This implies that  $\beta_{ca0} = 0$  and  $\beta_{mg0} = 0$ . Under this condition,  $m$  is always zero and the weighting function  $F$  is always one. And hence there is no change in the flow function despite the salinity or composition of the subsequent flood as confirmed by Lager[8].

The examples below use the simulation parameters in Table 2, which refer to the particular brine compositions given in Tables 3-5.

**Example Two:** Divalent cation in connate brine, but no low salinity induced desorption.

It is possible to have divalent ions in the connate brine, but have no desorption of these ions in the core if the composition and/or salinity of the invading brine does not support desorption of the ions. The following examples highlight this:

a) Ageing with connate water, flooding with sea water for 5 days, further flooding with low salinity sea water for 2 days and low salinity for another 3 days following the same sequence as reported in Skrettingland et al. [9]. The parameters used in this simulation are shown in Tables 2 and 3.

Result: After the seawater flooding, there was substantial desorption of divalent ions resulting in a change of  $F$ , from a value of 1 to 0.67 (Figure 5a). This change amounted to a change in the flow functions towards the improved flow functions. Subsequent flooding with low salinity seawater and low salinity water did not produce any further desorption of the divalent ions and hence same  $F$  of 0.67 and in turn no change in the flow function resulting in no substantial improved recovery with the low salinity floods (Figure 5b). This is the same trend that is seen in the corefloods reported by Skrettingland et al. [9], where low salinity seawater gave an increase by only 0.4-1.8% OOIP and low salinity water an increase of 0-2.8% OOIP for different reservoir cores at low pressure (6 bars).

b) Ageing with connate water, and flooding with a succession of seawater for 6 days, aquifer water for 2 days and low salinity water for 2 days following the sequence carried out by Alotaibi et al. [21]. The simulation parameters are listed in Tables 2 and 4.

Again the model results shows substantial desorption of divalent ions, after the seawater flooding, resulting in a change of  $F$  from a value of 1 to 0.92 (Figure 6a). Subsequent flooding with low salinity aquifer water and diluted aquifer water did not produce any further change in  $F$  and as a result no substantial improved recovery (Figure 6b). This is the same trend that is seen in the coreflood reported by Alotaibi et al. [21], where the aquifer water resulted in 2.2% additional oil recovery and no further recovery was seen with the diluted aquifer water.

c) Ageing with connate water, and flooding with a dilution of the connate water with a Berea core as used in Zhang and Morrow [22]. The simulation parameters are listed in Tables 2 and 5.

The result shows that flooding with the diluted formation water did not result in an appreciable desorption of the divalent ions and hence the weighting factor remained close to 1 resulting in no appreciable gain in recovery for the diluted connate water over the connate water (Figure 7). Zhang and Morrow [22] showed that Berea cores with similar property gave a recovery of about 70% OIIP with injection of connate water and most of the cores gave similar recovery with injection of diluted connate water.

**Example Three:** Divalent cation in connate brine plus subsequent low salinity induced desorption.

a) Ageing with connate water, and flooding with a succession of formation water, seawater, and aquifer water as done by Alotaibi et al. [21]. The simulation parameters are listed in Tables 2 and 4.

The recovery as shown in Figure 8, suggests that there is an improved recovery during the sea-water flood and further recovery during the low salinity aquifer flood. A similar result was seen by Alotaibi et al. [21] where seawater improved the recovery from 35% OIIP to 45% and low salinity aquifer flood improved recovery to 50%.

## CONCLUSIONS

The desorption of the divalent ions seems to play an important role in the low salinity water mechanism. We have calculated the amount of divalent ions that are attached to the core surface during waterfloods and shown that the absence of low salinity effect on some published data coincides with the NON-desorption of the divalent ions.

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Table 1 Brine compositions for static and dynamic experiments

| Salt                                 | Formation water (FW)<br>[g/l] | Sea water(SW)<br>[g/l] | Low salinity water (LSW)<br>[g/l] |
|--------------------------------------|-------------------------------|------------------------|-----------------------------------|
| NaCl                                 | 49.59                         | 23.38                  | 0.496                             |
| Na <sub>2</sub> SO <sub>4</sub>      | ---                           | 3.41                   | ---                               |
| NaHCO <sub>3</sub>                   | ---                           | 0.17                   | ---                               |
| KCl                                  | 1.04                          | 0.75                   | 0.010                             |
| MgCl <sub>2</sub> *6H <sub>2</sub> O | 2.22                          | 9.05                   | 0.022                             |
| CaCl <sub>2</sub> *2H <sub>2</sub> O | 3.74                          | 1.91                   | 0.037                             |



Table 2 Simulation parameters

|                                  | Example 2a | Example 2b | Example 2c | Example 3a |
|----------------------------------|------------|------------|------------|------------|
| Length of core                   | 10cm       | 50cm       | 7cm        | 12cm       |
| Mass of clay(Kg/bulk Litre Core) | 0.088      | 0.088      | 0.088      | 0.088      |
| CEC(moles/Kg of clay)            | 0.013      | 0.013      | 0.013      | 0.013      |
| Porosity                         | 0.274      | 0.274      | 0.274      | 0.274      |
| Absolute permeability            | 500md      | 150md      | 1100md     | 150md      |
| Oil Viscosity                    | 0.6cp      | 8cp        | 47cp       | 8cp        |
| Weighting constant, K            | 400        | 400        | 400        | 400        |
| Injection Velocity               | 0.01cm/day | 0.01cm/day | 0.01cm/day | 0.01cm/day |
| Rel. Permeability                | Figure 4   | Figure 4   | Figure 4   | Figure 4   |
| Brine Composition                | Table 3    | Table 4    | Table 5    | Table 4    |

Table 3 Brine composition\_ Example 2a after Skrettingland et al. [9]

| Ions                          | Connate water<br>[moles/litre] | Sea water<br>[moles/litre]] | Low salinity<br>Seawater | Low Salinity |
|-------------------------------|--------------------------------|-----------------------------|--------------------------|--------------|
| Na <sup>+</sup>               | 0.5282                         | 0.4651                      | 0.004651                 | 0.0085       |
| Cl <sup>-</sup>               | 0.5982                         | 0.5437                      | 0.005437                 | 0.0085       |
| Mg <sup>2+</sup>              | 0.0066                         | 0.0526                      | 0.000526                 | -            |
| Ca <sup>2+</sup>              | 0.0284                         | 0.0102                      | 0.000102                 | -            |
| SO <sub>4</sub> <sup>2-</sup> | -                              | 0.0235                      | 0.000235                 | -            |

Table 4 Brine composition\_ Example 2b and 3a after Alotaibi et al [21].

| Ions                          | Connate water<br>[moles/litre] | Sea water<br>[moles/litre] | aquifer<br>water | Diluted<br>aquifer water |
|-------------------------------|--------------------------------|----------------------------|------------------|--------------------------|
| Na <sup>+</sup>               | 2.3663                         | 0.7341                     | 0.0654           | 0.00654                  |
| Cl <sup>-</sup>               | 2.4245                         | 0.8807                     | 0.0727           | 0.00727                  |
| Mg <sup>2+</sup>              | 0.0066                         | 0.0938                     | 0.0027           | 0.00027                  |
| Ca <sup>2+</sup>              | 0.0264                         | 0.0166                     | 0.0098           | 0.00098                  |
| SO <sub>4</sub> <sup>2-</sup> | 0.0039                         | 0.0371                     | 0.0009           | 0.00009                  |

Table 5 Brine composition\_ Example 2c after Zhang and Morrow [22]

| Ions                          | Connate water [moles/litre] | Low salinity [moles/litre] |
|-------------------------------|-----------------------------|----------------------------|
| Na <sup>+</sup>               | 0.5931                      | 0.005931                   |
| Cl <sup>-</sup>               | 0.5480                      | 0.005480                   |
| Mg <sup>2+</sup>              | 0.0070                      | 0.000070                   |
| Ca <sup>2+</sup>              | 0.0190                      | 0.000190                   |
| SO <sub>4</sub> <sup>2-</sup> | 0.0486                      | 0.000486                   |

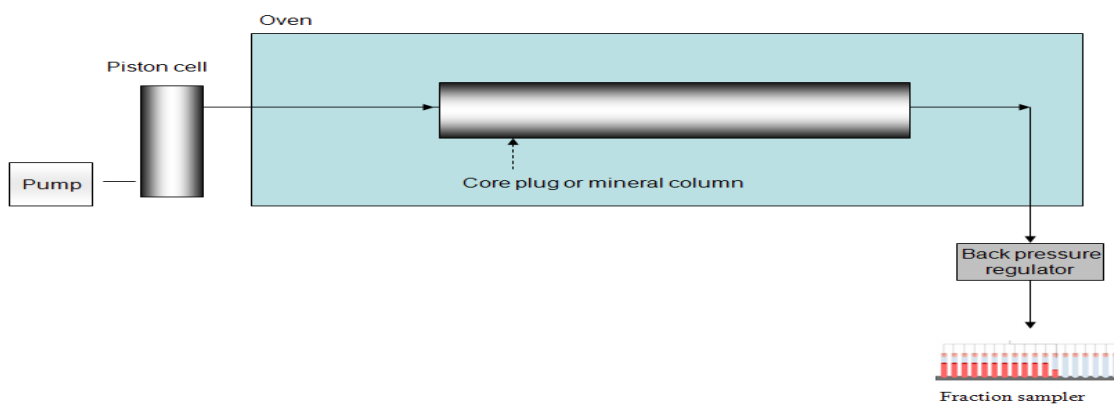


Figure 1. Flooding rig

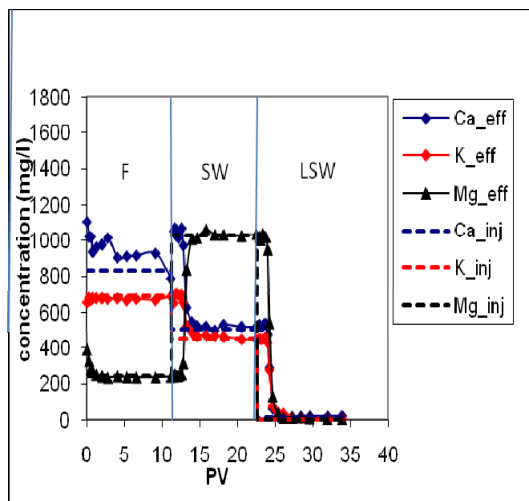


Figure 2a. Effluent ions from one phase water flood in Berea core. Injected concentrations are given as references.

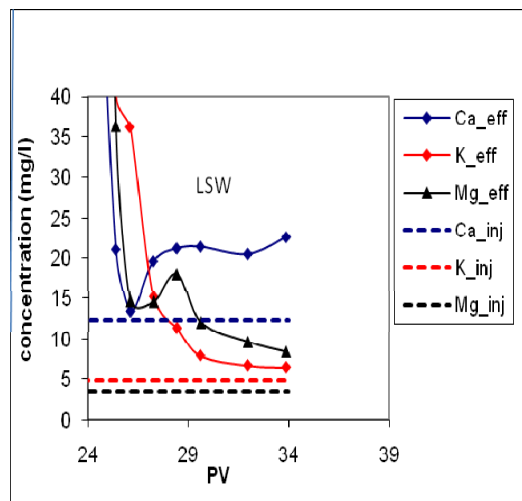


Figure 2b. Expanded LSW portion of Figure 2a

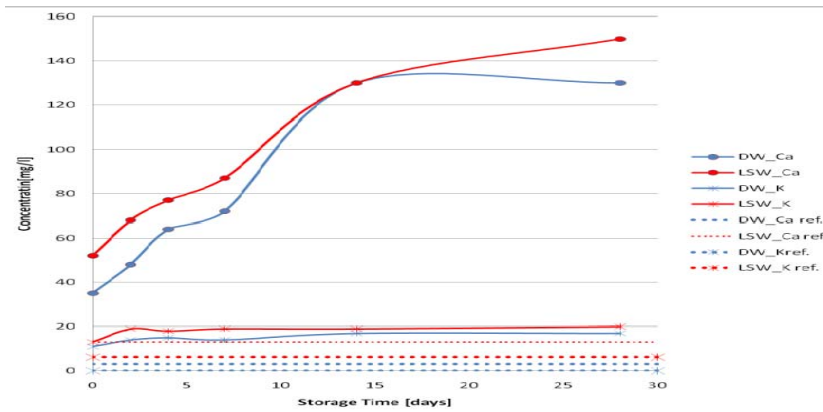


Figure 3. Ions from static experiment using reservoir rock mixed with low salinity water (LSW) and distilled water (DW). Concentrations of ions in waters are given as references

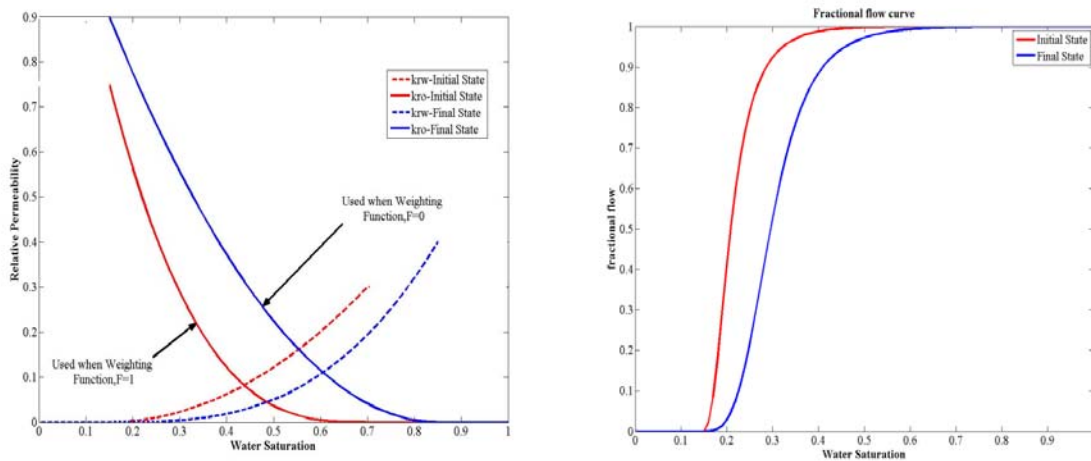


Figure 3. Showing the different flow functions, the red curves corresponding to the initial state when  $F=1$ , the blue curves for an extreme case when  $F=0$ . A linear interpolation between both curves is employed when  $0 < F < 1$

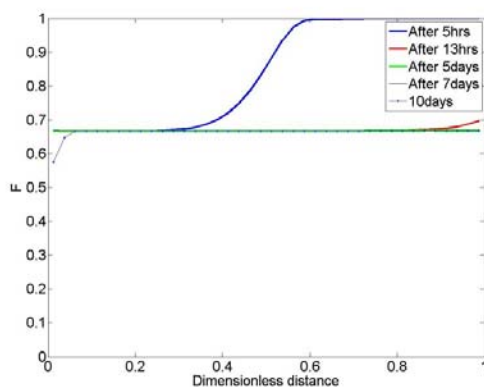


Figure 5a. Weighting function along the core at different times-Example 2a

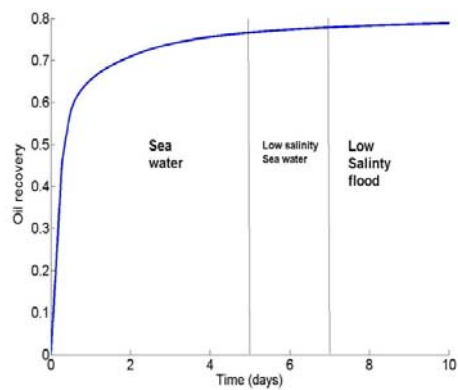


Figure 5b. Production curve Example 2a

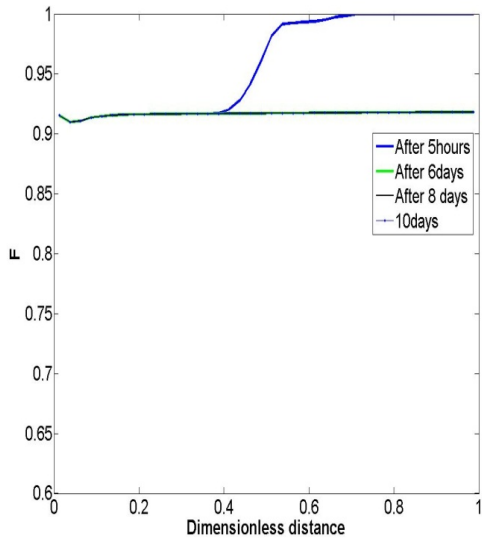


Figure 6a. Weighting function along the core at different times-Example 2b

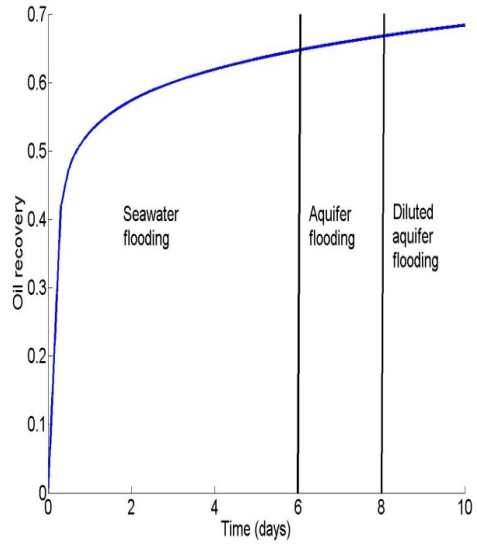


Figure 6b. Production curve Example 2b

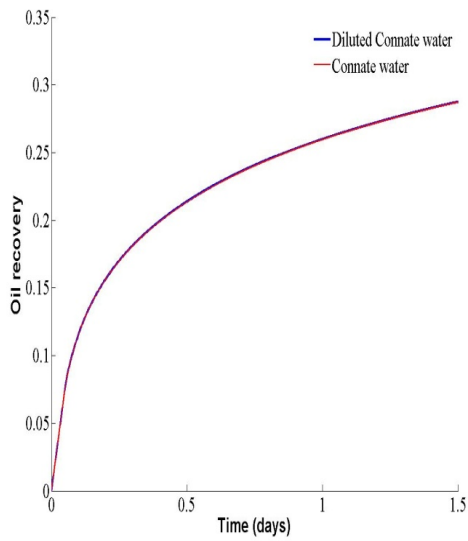


Figure 7. Recovery Example 2c

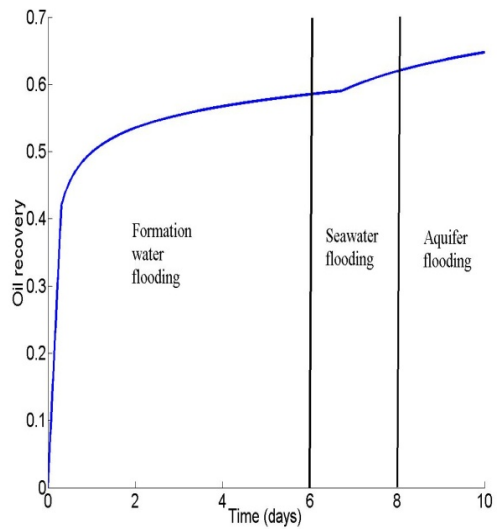


Figure 8. Recovery example 3a