

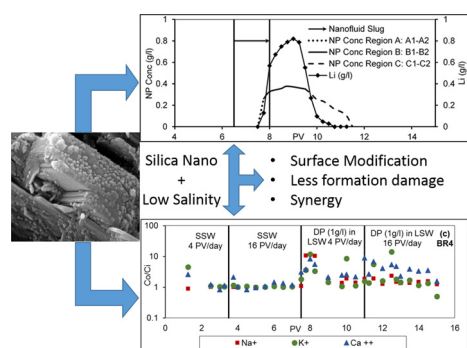
Adsorption of silica nanoparticles and its synergistic effect on fluid/rock interactions during low salinity flooding in sandstones

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GRAPHICAL ABSTRACT



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ABSTRACT

Several research works have shown the potential of incremental oil recovery by low salinity water (LSW) injection. The research in this area has also shown that LSW interaction with rock's mineral (sandstone and chalk) raises the potential for formation damage by produced fines. The objectives of this work are to address the adsorption of silica nanoparticles (NPs) on sandstone and their effect on fluid/rock interaction during LSW flooding. Isothermal static adsorption of NPs on sandstone mineral surfaces showed a higher adsorption affinity on quartz surface compared to kaolinite. This was also shown by scanning electron microscope images. The adsorption of NPs was enhanced by increasing salinity. To investigate the dynamic adsorption, a co-injection of about 0.033 g NPs slug with tracer (about 0.13 g of LiCl_2) as a reference. The estimated irreversible adsorption of NPs in the Berea flooding of core was about 35%. While estimated desorption of the flooded core was about 21.2%. Detailed mass balance analysis is included. It was observed that the adsorption/desorption processes of silica NPs are influenced by the pH wherein increased alkalinity favors NP desorption. NP adsorption on the mineral surface during combined LSW and NP flooding was shown to reduce mineral dissolution, ion exchange, loss of cementing mineral and reduced resistance to flow compared to LSW alone. Surface forces estimation showed that combining LSW with NPs reduced the repulsion between fines and Berea. The work here demonstrated the synergistic effect of combining the two technologies of LSW and nanoparticles where the probability of formation damage in sandstone reservoirs is reduced.

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1. Introduction

Use of nanoparticles (NPs) has emerged as an Enhanced Oil Recovery (EOR) technique during the past decade. Nanofluids (NF) which are dispersions of NPs typically under the size of 100 nm in a base fluid have been studied as an injection fluid for improving oil recovery from petroleum reservoirs. The main advantage of NPs is their size and high surface area which allows them to pass through the pore network in the reservoirs and be effective at relatively low volume concentration as compared to other EOR agents [1]. Among the various type of NPs, special attention has been paid to silica NPs due to their hydrophilic nature and ease of surface functionalization [2]. Silica NPs can alter the wettability of the oil wet rock surface towards more water wet and this has been attributed and studied as the main mechanism that improves recovery due to application of Silica NPs [3–6]. In addition, core flood studies conducted by different research groups have shown the silica NPs can increase recovery in sandstone reservoirs [3,7–10]. Another popular EOR technique for sandstone reservoirs is use of low salinity water injection [11–13]. This techniques generally involves altering or lowering the salinity to injection brines. However, lowering the salinity of injection brine can have detrimental effects. Khilar and Fogler [14] identified the existence of a Critical Salt Concentration (CSC) for permeating fluids in Berea sandstones below which clay particles get released and cause formation damage. Formation damage by lowering brine salinity has also been reported and studied by other researchers [15–17] and thus choosing optimum brine salinity in low salinity projects is limited by the CSC [18]. Thus fluid/rock interactions are very important during low salinity flooding. Arab and Pourafshary [18] investigated different NPs as surface modifiers by soaking the porous medium in NFs and then testing the ability of the modified porous medium to hinder the transport of artificial fines in water saturated porous medium. They suggest that combining low salinity and with NPs may help overcome the detrimental effects of formation damage associated with low salinity flooding. The current study investigates the adsorption of silica NPs on sandstone minerals and its effect on fluid/rock interactions during oil recovery by low salinity flooding. It has been shown that the adsorption of silica NPs on sandstone mineral surface can reduce mineral dissolution and formation damage in Berea sandstones.

2. Materials and methods

The Silica NPs (DP 9711) used in this study were provided by Nyalco Nanotechnologies. The NPs were obtained at 30% wt. concentration, dispersed in Deionized Water (DIW) and pH 3. For ease, these NPs are referred to as DP in this study. The NPs are spherical and surface functionalized with a proprietary coating. The NPs have an average particle size of 20 nm as claimed by the manufacturer. The NFs used in this study were prepared from the stock fluid by diluting it with appropriate brines. Berea outcrop cores were used as the porous media. The mineral composition of the used cores is listed in Table 1. Analysis grade quartz and kaolinite mineral powders were acquired from Sigma–Aldrich with chemical compositions: SiO_2 and $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$, respectively. The specific surface area of the used mineral powders are $0.62 \text{ m}^2/\text{g}$ and $8.56 \text{ m}^2/\text{g}$, respectively which has been calculated previously by the water adsorption isotherm [19]. The model oil used in

Table 1
Mineral composition of Berea sandstone cores.

Mineral	Semi-quantitative (%)
Quartz	94
Kaolinite	1
Muscovite	1
Microcline	1

Table 2
Synthetic oil properties.

Temperature °C	Viscosity (cP)	Density (g/ml)
20	0.92	0.73
50	0.5802	0.7683
70	0.4812	0.7525

this study was n-decane acquired from Merck. N,N-Dimethyldodecylamine (NN-DMDA) was added to n-decane at a concentration of 0.01 mol/l to prepare the synthetic oil. The properties of the synthetic oil (estimated from PVT Sim) are listed in Table 2.

Zetasizer Nano ZSP from Malvern instruments was used to characterize the average size and zeta potential of the NPs. Scanning electron microscopy was performed on a Supra 35 V P SEM with an integrated EDXRF analyzer to visualize the adsorption of the NPs on the Berea core pieces treated with NF. NP concentration during static isothermal adsorption tests and in the effluents from core floods were determined using a dual beam UV–vis 1700 spectrophotometer from Shimadzu Corporation. The schematic of the core flooding setup used in this study is shown in Fig. 1. The concentration of cations in effluents produced from core floodings was determined by a Dionex ICS-5000 Ion Chromatograph (IC) from Thermo Fisher Scientific.

2.1. Brines and nanofluid

Synthetic seawater (SSW) and Low Salinity Water (LSW) at 1:10 dilution of SSW were the used brines. Their ionic compositions are listed in Table 3. The particles size (average hydrodynamic diameters) and zeta potential exhibited by the NPs in NFs prepared in LSW and SSW are listed in Table 4. At the NPs concentration (1 g/l) used in this study, we did not observe any aggregation behavior. Particle size measurements made after 3 months of nanofluid preparation were within $\pm 5 \text{ nm}$ of the original measurements for all nanofluids including in seawater (high salinity). Griffith, Ahmad, Daigle and Huh [20] made a similar observation (by observing particle size with time) only for very high concentration (200 g/l) in high salinity (20 wt.% API brine).

2.2. Adsorption of NPs on minerals

The adsorption behaviour of the NPs on the mineral surfaces in sandstones were investigated by two approaches: (1) static isothermal adsorption on individual mineral powders and (2) dynamic adsorption of NPs in Berea core during low salinity flooding. A series of batch adsorption experiments were performed at room temperature to study the static isothermal adsorption of the used NPs on quartz and kaolinite mineral surface. The experiments were performed in DIW and SSW as the media to address the effect of salinity on NPs' adsorption. 0.15 g of mineral was added to NF prepared at a particular concentration and salinity. This fluid was then agitated (in a rotary agitator) for 24 h. Thereafter, the minerals were removed from the fluid. The remained concentration of NPs in the fluid was determined by measuring its absorption in a dual beam spectrophotometer at 240 nm wavelength, comparing it with the constructed calibration curve and making baseline corrections for the contribution of minerals [21].

The dynamic adsorption of NPs was addressed by injecting a slug of NPs with into a Berea core. A dried Berea core was vacuum saturated with LSW and loaded in to the core holder (Fig. 1). Confining pressure of 25 bar was applied on the core and the injection of the fluid was performed at a constant flow rate of 10 pore volumes (PV)/day at room temperature. The details of the core used is listed in Table 5. Multiple PV of LSW was injected into the core. Thereafter, 1.5 PV slug of NF (1 g/l DP in LSW + 0.1 mol/l LiCl tracer) was injected into the core followed by post flush with LSW. The produced effluents were collected at

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