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Prediction of Critical pH for Fines Migration Pre- and Post-Nanofluid Treatment in Sandstone Reservoirs using the DLVO Modelling

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Abstract - Injection water pH affects the release of fines in sandstones. The force equilibrium between fines and sand governs the attachment or release of fines in the system. At a pH higher than a critical value, fines are released and block the pores, causing formation damage. The fines release can be avoided by adjusting the pH and using nanofluids. This paper introduces the concept of DLVO modelling to estimate the critical pH before and after the application of nanofluids without extensive experimentation. Scanning electron microscopy determines the average size of in-situ fines collected from sandstone core. Injection brine of 11700ppm and 0.1wt% nanofluid are prepared, zeta potentials of dispersed sand are measured with varying pH from 2 to 12, and the resulting attractive and repulsive surface forces between fines and sand grains are quantified. The DLVO models are developed to predict the mobilization of fines and a critical pH before and after the application of silica nanofluids. The zeta potentials are measured by a Zetasizer and are in the range of -5 mV (less repulsion) to -31 mV (more repulsion). Furthermore, the application of nanofluids increases the zeta potential to a range of -3 mV to -24.9 mV, indicating a compression in electric double layers. Measured zeta potentials, ionic strength, and fine size are used as inputs to compute surface forces, and DLVO models are developed. The critical pH, at which total DLVO interactions shift from negative to positive, as predicted by the model, is about 8. The DLVO model also predicted an improved critical pH of 11 following the use of nanofluids, demonstrating a reduction in repulsion forces. DLVO modelling approach helps estimate a critical pH before and after applying nanofluids, and nanotechnology validates nanoparticles' ability to control fines migration and improve critical pH for waterflooding and alkaline flooding operations.

***Keywords*:** sandstone, formation damage, fines migration, silica nanoparticles, zeta potential, DLVO modelling, pH

1. Introduction

Oil-containing sandstone reservoirs are a significant contributor to global energy production and account for approximately 60 % of the world's petroleum reservoirs [1]. Sandstone reservoirs have ultra-fine quartz and clay particles attached to the sand surface, and this attachment creates a sand-fine-brine (SFB) system [2]. Clay minerals, such as kaolinite, illite, montmorillonite, and chlorite, are the ones that have the widest distribution and are responsible for the generation of the most migrating fines [3], [4]. Electrostatic and gravitational forces hold fine particles to the rock surface. The electric double layers (EDLs) around fine particles and sand grains are compressed in the SFB system due to the high charge density, maintaining the fine particles in equilibrium with sand grains in high salinity formation water (FW) [5].

In an environment with a high salinity formation of water, there are electrostatic forces of van der Waals attraction (P_{VAW}), electric double layer repulsion (P_{EDL}), and weak Born repulsion (P_{Born}) between fine particles and sand grains. Fine particles are affixed to the sand's surface by the overall attractive interaction energy of the SFB system. Fine particles remain attached to sand grains mainly due to the van der Waals force, and the strength of this force is proportional to the fine particle size as well as the distance from the surface of the sand that separates them. The electric double layer repulsion force, on the other hand, tends to detach fine particles from the rock surface, and the zeta potential of the SFB system is the primary factor that determines the strength of this potential [6]. Derjaguin, Landau, Verwey, and Overbeek (DLVO) proposed a theory based on surface force quantification and analysis that can be used to model the dynamic behaviour of a sand-fine-brine system and investigate the effect of various crucial factors on fine migration initiation [7]–[9]. Equation 1 illustrates the DLVO-based total potential (P_{Total}) of a system that consists of a fine particle and a sand grain surface. This equation represents a summation of all the energies that are present in the SFB system.

$$P_{Total} = P_{VAW} + P_{EDL} + P_{Born} \tag{1}$$

Fines can be released and migrate under certain conditions, and their movement can be predicted as a function of the injected brine's salinity, ionic strength, flow rate, pH, and presence of monovalent and divalent ions, as well as the concentration of in-situ fines and temperature. Because of low salinity waterflooding, as the SFB system salinity decreases due to the injection of low salinity water, electric double layers around fines and sand grains expand, and fine particles are dislodged due to high repulsion forces, and migration begins inside the reservoir even at low flow rates. Detached and migrating fines might plug pore throats, causing productivity and injectivity issues and a high-pressure drop in the system [10]–[12].

The pH of the SFB system may rise after water injection due to the solubilization and dissolution of rock minerals such as calcite and siderite. This has a considerable impact, particularly in the process of water flooding, on the release of fine particles in sandstones above a specific pH value called a critical pH. The release of fines because of an increase in the system pH can be attributed to an increase in the repulsive forces and modelled using the DLVO approach [13].

In our previous studies, DLVO models were developed, and critical salt concentrations were estimated based on the characterization of surface forces between fines and sand grains under low salinity water injection with monovalent and divalent ions [6], [14]–[16]. The application of silica nanoparticles in the form of nanofluid provides promising results in controlling fines migration and reducing the critical salt concentration. The DLVO models also incorporated the effects of silica nanoparticles, and all model results were validated experimentally. The current research provides a new insight into the estimation of a critical pH using the DLVO modelling technique and incorporates the effect of silica nanoparticles in the model to mitigate fines migration and improve the critical pH value.

2. Research Methodology

2.1. Brines

In the experimental phase, high salinity formation water (FW) of 77000 ppm was employed to saturate the core samples to recreate sandstone reservoir conditions. NaCl brines of 11693 ppm with a pH range of 7-12 were used to determine the critical pH for the given SFB system. The ionic compositions of formation water and base brine are given in Table 1.

Table 1: Composition of formation water and NaCl brines.

Ions	FW (ppm)	0.2M NaCl (ppm)
Na^{+}	23426	4600
Ca^{2+}	4448	-
Mg^{2+}	1300	-
Cl^{-}	47781	7093
Total	76955	11693

2.2. Nanofluid

To investigate the effect of nanoparticles on critical pH value, Glantreo Ltd. Ireland provided silica nanoparticles of 20 nm size in the form of a 25 wt% concentrated solution. These nanoparticles were chosen because of their hydrophilicity, better stability, low toxicity, and superior performance in controlling fines migration in sandstone core samples. Furthermore, 0.1 wt% nanofluids were produced by diluting the dispersion with 0.2M NaCl brine.

2.3. Zeta Potential Measurements

DLVO modelling requires sand-brine and sand-nanofluid zeta potentials as inputs. A sandstone core sample was crushed in a cone crusher and sieved to a 40-micrometer sand particle size for this purpose. To eliminate any contaminants on the sand grains, the sieved sample was washed with distilled water, HCl, acetone, and finally distilled water. With an ultrasonic homogenizer, the crushed sand particles were dispersed in brine and nanofluid, and the zeta potentials were evaluated with varied pH using Malvern Zetasizer (NanoZS) and a Titrator system.

2.4. Characterization of Fine Particles

The sandstone core was used to collect the in-situ fines, and scanning electron microscopy (SEM) was utilized to determine the morphology and average size of the fine particles. In addition, SEM-EDS analysis was carried out to collect data on the chemical composition of fines.

2.5. DLVO Modelling

The average size of fine particles, the ionic strength of the brine, and measured zeta potentials at different pH were utilized as inputs for DLVO modelling, and surface forces were computed using available methods indicated in our previous research [6]. The DLVO model was used to predict the critical pH, at which the total interaction energy shifted from negative to positive. The model was also utilized to test the effectiveness of silica nanofluid in changing surface forces and reducing fines migration even at higher pH levels.

3. Results and Discussion

3.1. Zeta Potential Results

Zeta potentials of dispersed sand particles were measured before and after the application of silica nanoparticles as shown in Fig. 1. As the pH of the solution increases, the electric double layer expands, and the corresponding zeta potential decreases. On the other hand, the application of silica nanoparticles decreases the expansion of the electric double layer and increases zeta potential.

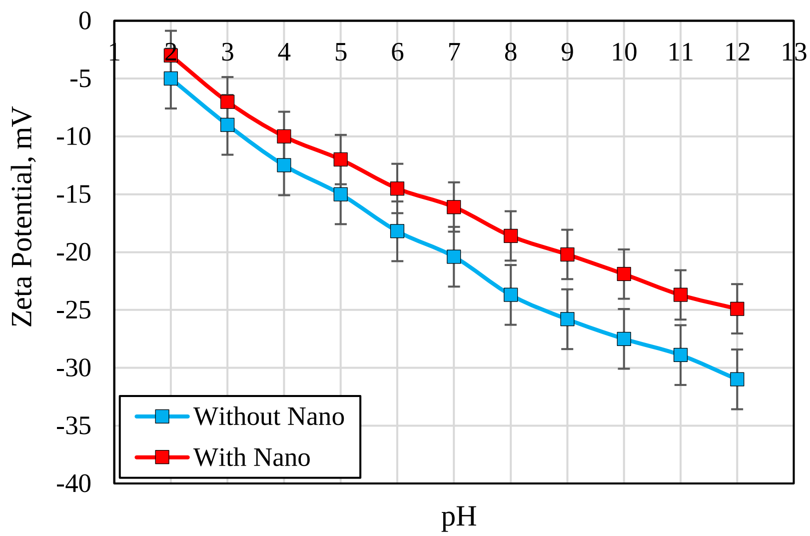

Fig. 1: Zeta potentials before and after the application of silica nanoparticles.

3.2. Fines Characterization

SEM and SEM-EDS were used to examine the collected fines. The fine particles were mostly kaolinite clay ($Al_2Si_2O_5(OH)_4$), according to SEM-EDS and X-ray diffraction (XRD) analyses, and their average size was around 800-900 nm.

3.3. Critical pH by DLVO Modelling

Surface forces between fine and sand grain were estimated using observed zeta potentials under various pH conditions without nanoparticles, brine ionic strength, and average fine particle size, and then the total interaction energy of each system was determined using DLVO modelling, as shown in Fig. 2. The sum of attraction and repulsion potentials is the total interaction energy of the system under consideration. Because of compacted electric double layers around fines and sand grains in a low pH environment, such as a pH=3 solution, forces of attraction dominate repulsion forces, and the total energy is negative, indicating that there will be no fines migration in the system. The same findings were achieved for the total interaction energy in the negative region for pH 4-7 environments. However, for brine of pH=8, the total interaction energy shifted from negative to positive indicating the dominance of repulsive forces in the system at a separation distance of around 0.25 nm. This can be attributed to the expansion of electric double layers around fines and sand grains under high pH conditions. Based on the DLVO model results, pH 8 was predicted to be the critical pH, and fines migration in the SFB system may occur at or above this value.

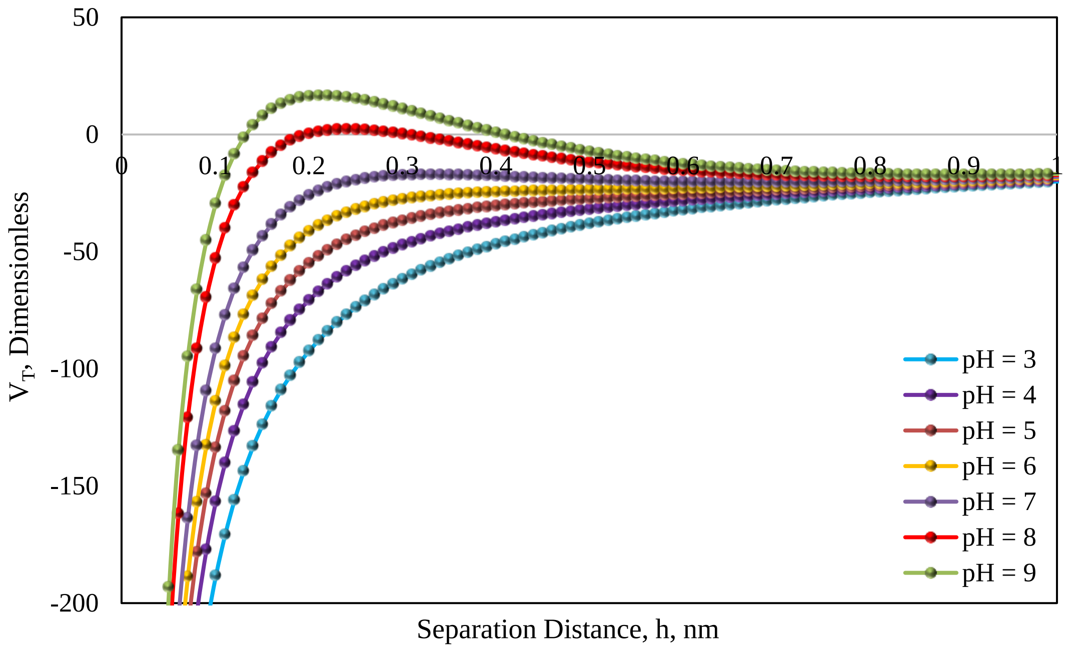

Fig. 2: Prediction of critical pH using DLVO modelling.

3.4. Effect of Nanoparticles on Critical pH

Similarly, the surface forces between fines and sand grains were quantified using zeta potentials under different pH conditions with the utilization of silica nanoparticles, brine ionic strength, and average fine particle size, and the total interaction energy of each system was calculated using DLVO modelling, as shown in Figure 3. By altering the surface charge of sand grains, silica nanoparticles assisted to limit the expansion of electric double layers in high pH environments. Forces of attraction dominate repulsion forces in the presence of compacted electric double layers around fines and sand grains in the pH range of 6-10, and the total energy is negative without any fines migration in the system. The overall interaction energy shifted from negative to positive for brine with pH=11, suggesting the dominance of repulsive forces in the system. This occurs because of nanoparticles' adsorption on the surface of the sand grains, which causes the electric double layers to contract even under higher pH values. The results of the DLVO model indicated that pH 11 would be the new critical pH, and it was projected that fines migration in the SFB system would take place at this value or higher after the application of silica nanoparticles.

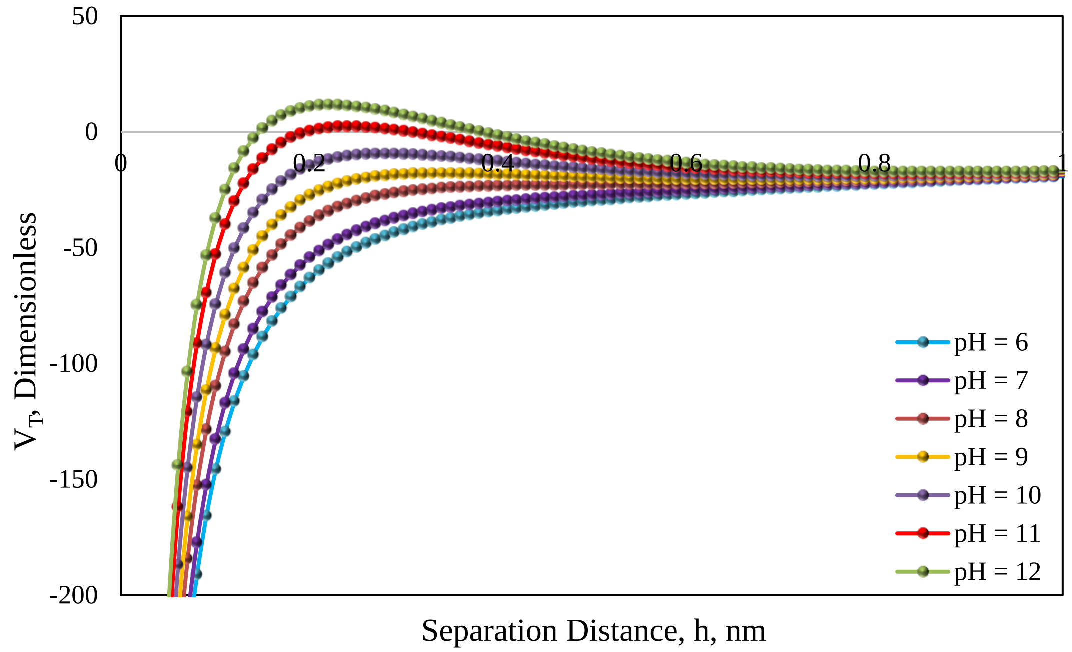

Fig. 3: Prediction of a new critical pH after the application of nanoparticles.

4. Conclusions

During waterflooding and alkaline flooding operations, estimating the critical pH of injection water is a crucial factor to prevent formation damage due to fines detachment and migration. The DLVO modelling approach, which is based on the quantification and analysis of surface forces, is a useful tool for analyzing fines migration and control in sandstones during alkaline flooding without having to conduct major experiments. The following is a summary of the results:

- A critical pH for a sand-fine-brine system was predicted using the DLVO modelling technique.
- The DLVO model predicted a critical pH of 8 at which the total interaction energy of the SFB system moved from negative to positive, indicating the dominance of repulsion forces in the system.
- In addition, after the application of silica nanoparticles, the DLVO model predicted a new critical pH of 11.
- An increased critical pH value indicates the effectiveness of silica nanoparticles in controlling fines migration and avoiding formation damage even at a higher pH during alkaline flooding operations.

Acknowledgments

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