Fine Migration Control in Sandstones: Surface Force Analysis and Application of DLVO Theory

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ABSTRACT: Formation damage caused by fine migration and straining is a well-documented phenomenon in sandstone reservoirs. Fine migration and the associated permeability decline have been observed in various experimental studies, and this phenomenon has been broadly explained by the analysis of surface forces between fines and sand grains. The Derjaguin–Landau–Verwey–Overbeek (DLVO) theory is a useful tool to help understand and model the fine release, migration, and control phenomena within porous media by quantifying the total interaction energy of the fine–brine–rock (FBR) system. Fine migration is mainly caused by changes in the attractive and repulsive surface forces, which are triggered by mud invasion during drilling activity, the utilization of completion fluid, acidizing treatment, and water injection into the reservoir during secondary and tertiary recovery operations. Increasing pH and decreasing water salinity collectively affect the attractive and repulsive forces and, at a specific value of pH, and critical salt concentration (CSC), the total interaction energy of the FBR system (Vf) shifts from negative to positive, indicating the initiation of fine release. Maintaining the system pH, setting the salinity above the CSC, tuning the ionic composition of injected water, and using nanoparticles (NPs) are practical options to control fine migration. DLVO modeling elucidates the total interaction energy between fines and sand grains based on the calculation of surface forces of the system. In this context, zeta potential is an important indicator of an increase or decrease in repulsive forces. Using available data, two correlations have been developed to calculate the zeta potential for sandstone reservoirs in high- and low-salinity environments and validated with experimental values. Based on surface force analysis, the CSC is predicted by the DLVO model; it is in close agreement with the experimental value from the literature. The critical pH value is also estimated for alkaline flooding. Model results confirm that the application of NPs and the presence of divalent ions increase the attractive force and help to mitigate the fine migration problem. Hence, a new insight into the analysis of quantified surface forces is presented in current research work by the practical application of the DLVO theory to model fine migration initiation under the influence of injection water chemistry.

INTRODUCTION

The decrease in ionic strength (I) and change in ionic composition of formation water lead to fine migration and permeability reduction in subsurface porous and permeable sandstone reservoirs containing various types of clay minerals. Such minerals are present in the pore space as either agglomerate particles or as fine particles which cover the sand grains. Clay minerals are alumino-silicates with a very specific layered structure. The basic building blocks of clay minerals are layers of silica, alumina, and magnesia. There are major three types of clay minerals which include kaolinite, montmorillonite, and illite/mica. Sandstones consist of a matrix of quartz grains enclosing an interconnected pore space. The nature of the depositional environment can lead to the presence of siliceous fines because of grain compaction and crushing because of applied stresses as well as fine deposition. These fines are generally held within the formation water film that surrounds the quartz grain in water-wet conditions. Fine migration may happen in different types of natural and technical processes, such as water aquifer recharging by some external water source, underground formation water disposal, groundwater flows, and invasion of drilling muds, invasion of completion fluids, acidizing and waterflooding treatments, high rate oil and gas production and injection, and improper design of oil recovery processes with low-salinity water injection.4–15 Water injection operations into sandstone and carbonate reservoirs performed by reducing the salinity and tuning the ionic composition are a promising and evolving technology to...
maximize oil recovery, primarily by modifying the wettability of the crude—brine—rock system. During the aforementioned process of flooding, the salinity, chemistry, and injection rate of injected brine play a vital role in altering the rock wettability and changing the surface forces between fines and sand grains in sandstone reservoirs, which affect the efficiency of the procedure. Fine particles can detach, become suspended in the injected fluid, and form a colloidal system in the reservoir because of the alteration of attractive and repulsive surface forces; while they move with the injected fluid/brine, they may block the pore throats. This phenomenon is referred to as straining: it blocks already open pores and results in formation damage, with a substantial decline in formation permeability as shown in Figure 1.

![Figure 1. Migration of natural fine particles in the reservoir (reproduced with permission from J. Nat. Gas Sci. Eng. 2020, 73, 103047).](image)

Fine migration in sandstone reservoirs is supposed to be one of the possible mechanisms of enhanced oil recovery (EOR) in low-salinity projects. It provides better mobility control by reducing because of clay swelling. Later, Khalir and Fogle found that when injected fluid salinity falls below a critical salt concentration (CSC), fines are released and migration starts within porous media. Fine migration and subsequent permeability reduction were also confirmed by experimental research. Kumar et al. and Mansouri et al. used scanning electron microscopy (SEM), the field emission SEM, and atomic force microscopy to visually show the mobilization of mixed-wet kaolinite particles with high-resolution images.

Attractive forces are responsible for retaining fines on the rock surface, whereas repulsive forces try to detach fines and promote migration. The Derjaguin–Landau–Verwey–Overbeek (DLVO) model incorporates surface forces and calculates the total interaction energy for the system, which is either positive or negative based on the contribution of each energy component.

In this paper, surface forces are quantified, and the DLVO model is used to predict the CSC for NaCl and to estimate a critical pH value for alkaline flooding. The model confirms that NPs increase the attractive energy and help fixate fines on sand grains. Furthermore, this tool has estimated an even lower CSC if there are divalent ions in the solution, which suggests the idea of tuning/adjusting the ion composition of injection water to avoid fine migration. Two correlations for the zeta potential calculation have also been developed and validated using further modeling. Therefore, the present study concludes that the analysis of quantified surface forces combined with DLVO modeling is a powerful tool to predict and control fine migration in porous media, and further research on the sensitivity of important parameters can improve the results.

## FINE—BRINE—ROCK SYSTEM

The detachment of fine particles from the sand grain surface is the initial step in the process of fine migration in sandstones. A comprehensive understanding of this detachment process is necessary to analyze conditions for the migration and the resultant formation damage. Generally, two types of forces are responsible for the detachment and mobilization of fine particles. These forces are classified as colloidal forces and hydrodynamic forces. Colloidal forces are electrostatic in nature, and they are further divided into two types, which are London dispersion forces and electrical double-layer repulsive forces between particles. The hydrodynamic forces are mainly related to the flow of permeating fluid through porous media. Synthetic fines and silica glass beads have
been used in numerous studies to mimic sandstone reservoirs with fine particles, as shown in Figure 2, because they are spherical. However, kaolinite in sandstone reservoirs has a platelet structure with a finite thickness, while natural sand and glass beads both have infinite thicknesses (IT) as compared to fine particle size. Based on the aforementioned configuration of synthetic/natural fines and sand/glass beads, generally, there are different electrostatic energies for two different systems: the sphere–IT plate and the kaolinite platelet–IT plate.

An analysis of a single fine particle of presumably spherical shape on a sand grain surface had been performed to describe the conditions required for the detachment of fine from a flat surface. Figure 3 describes the sphere–IT plate model designed to mimic a spherical fine particle attached to the pore/rock grain through which a high-salinity permeating liquid is flowing. However, Figure 4 demonstrates a kaolinite platelets–IT plate configuration, where small kaolinite platelets are present on a sandstone grain. Most of these platelets are in the form of clusters and can move together based on attractive forces between individual plates. Similar kaolinite platelet configurations have also been found in other sources.

The separation distance (h) between a fine particle and the pore surface in Figure 3 is quite small (usually on the order of 10⁻¹ nm), and additionally, these fine particles are subjected to the hydrodynamic forces of the flowing liquid during production and injection processes. There are different energy contributions from colloidal and hydrodynamic forces, and the total energy of all interactions between a fine particle and the pore/grain surface must be determined precisely in the DLVO model to incorporate the effects of attractive and repulsive forces. If the net interaction energy of the system comes out to be positive, it means repulsive forces have dominance over the attractive forces; as a result, fines will be detached from the surface, and migration will start in the porous medium.

During early research studies and to date, a single fine particle of spherical shape on a sand grain flat surface (sphere–plate model) has been extensively used for the calculation of DLVO interactions because of the simplicity of the approach. A few researchers have utilized a plate–plate model for the quantification of interaction energies. The single sphere model can be accurately used for synthetic fines and glass bead configurations, but when it comes to natural kaolinite and sand grain configurations, it can provide erroneous results because natural kaolinite has a platelet structure and must be modeled with a kaolinite platelets–IT plate model. In some studies during the last few years, clustered fine particles’ detachment and combined movement were assumed instead of a single fine particle model. Recently, Chequer et al. used this new idea to show that the single-colloid single-surface system is not an accurate representation of colloidal behavior in porous media and significantly underestimates the critical velocity of the fluid to initiate the fine migration. Experimental results were in close agreement with the clustered fines model.

### DLVO Theory

The well-known DLVO theory was established by Derjaguin, Landau, Verwey, and Overbeek. It describes the V of the system incorporating attractive and repulsive forces because of the van der Waals attractive potential (V_{LVW}), electric double-layer (EDL) potential (V_{EDL}), and Born repulsive potential (V_{BR}). This theory assumes that the V_{LVW}, V_{EDL}, and V_{BR} potentials are independent of each other and therefore can be added for the particle–plate system, using either a sphere–plate or plate–plate model configuration, to quantify total interaction energy at each interacting distance. The DLVO-based V of the system composed of a fine particle and a pore surface is presented as eq 1.

\[
V_T = V_{LVW} + V_{EDL} + V_{BR}
\]  

Generally, the DLVO theory provides good estimates for the surface–surface forces with a separation distance of around 5 nm, provided that all the important parameters, such as the particle size, I, Hamaker constant (A_H), and zeta potential (ζ), are accurately and precisely measured. The total energy of a
The total energy was attractive after the application of NPs as DLVO sphere experimental results, they also measured the zeta potentials of experiments on glass beads to mitigate the mechanism of coal purification and coal particles was repulsive after the adsorption of the experiments using SiO2 and MgO NPs in the pre-oscillating dipoles of atoms. These forces are weak chemical association with any type of ionic or covalent bonds. It decays in forces, but still play a critical role when colloidal particles are migration problem, computed total interactions, and found that MgO NPs improved the attractive forces between fines and the grain surface. Arab and Pourafshary performed several experiments on glass beads to mitigate the fine migration issue, accompanied by low-salinity flooding. They used five different types of NPs to control fine migration and found that ZnO and Al2O3 NPs were the best at this task. In addition to experimental results, they also measured the zeta potentials of the system before and after the application of NFs and applied a DLVO sphere—IT plate model to calculate interactive energy. The total energy was attractive after the application of NPs as compared to the nontreated case. Arab et al. performed experiments using SiO2 and MgO NPs in the preflush mode and found that 0.03% MgO NPs performed the best among all scenarios. They confirmed their experimental findings with the DLVO theory by calculating the total interaction potential between the rock and fine particles.

Assef et al. demonstrated their work to mitigate colloidal particle movement in porous media by using MgO NPs, and 97% retention of fines was observed. They utilized the extended DLVO (X-DLVO) theory by incorporating the effect of acid–base energy and neglecting the hydrodynamic forces and quantified the total interaction energy of the system. Zou et al. applied the X-DLVO theory to investigate the adsorption of anionic polyacrylamide onto coal and kaolinite particles. Based on the results, they observed that the VIT between kaolinite and coal particles was repulsive after the adsorption of the polymer on coal particles, which proves the effectiveness of the mechanism of coal purification.

**QUANTIFICATION OF INTERACTION ENERGIES**

**London–Van Der Waals (V_{LVW}) Interaction Energy.** In particle physics, there exists an attractive force between similar particles/plates when they are infinitesimally close to each other. A German-American physicist, Fritz London, published the first satisfactory microscopic theory of dipole–dipole dispersion forces. This attractive force is a distance-dependent force between molecules, atoms, and particles and does not have any association with any type of ionic or covalent bonds. It decays slowly and acts at a distance less than 10 nm. The main cause of this electrostatic force is the presence of permanent and oscillating dipoles of atoms. These forces are weak chemical forces, but still play a critical role when colloidal particles are infinitesimally close to each other in a solution. Based on the sphere—IT plate model, London–van der Waals energy (V_{LVW}) is presented in two forms, in eqs 2, and 3:

\[
V_{LVW} = -\frac{A_H a_p}{12h}
\]  

Chequer et al. presented a new model based on the clustered fine sphere–IT plate model, as shown in eq 4.

\[
V_{LVW} = -\frac{A_H}{12\pi\varepsilon}
\]  

The negative sign of V_{LVW} demonstrates the attractive nature of this potential.

**EDL (V_{EDL}) Interaction Energy.** When charged colloidal particles (fines) are immersed in an electrolyte solution of specific ionic strength, mobile ions from the electrolyte solution form an ionic film around the particles. Based on the positive or negative charge of a particle, oppositely charged ions from the surrounding electrolyte solution are attracted and form an ionic layer over the charged particle called a compact layer (stern layer), which is moved with the particle. The excess charge on the compact layer is balanced by the oppositely charged ions from the electrolyte solution forming another layer, which is called the diffuse layer (slipping plane). In the diffuse layer, ions are not tightly bound to each other and are free to move to and from the electrolyte solution. These two layers are electrostatic, and their combined effect is called the EDL, as shown in Figure 5. The potential difference between these two layers is called zeta potential and is denoted by ζ. Zeta potential provides the closest surface potential estimate and is used in the quantification of the EDL interaction energy. Zeta potential is not directly measured and is obtained by applying an electric field across the dispersion: this process is called electrophoresis. Particles within the dispersion with a specific zeta potential value move toward the electrode of opposite charge with a velocity proportional to the magnitude of the zeta potential. At lower ion strength, such as in the low-salinity

![Diagram of EDL and corresponding zeta potential](reproduced with permission from Elsevier: Amsterdam, The Netherlands, 1995; pp 113–156).
injection condition, EDLs that have already formed around the sand grain surface and the fine particles expand and overlap, which leads to a repulsive interaction energy ($V_{EDL}$). High repulsive force may detach the fine particles from the sand grain surface. The repulsive force is higher at a lower solution salinity.

Regarding the formulation of $V_{EDL}$, the simplest case of sphere–plate geometry is used, as shown in Figure 3. For the boundary conditions, the rock and fine particle surfaces both may have a constant surface potential or constant charge, or one of the surfaces may maintain its charge density constant while the other surface possesses a constant potential (mixed case). Generally, fine migration can be modeled with a constant potential case because zeta potential is easy to measure as compared to surface charge. $V_{EDL}$ can be calculated by different formulae, such as eqs 6, 7, 109 or 8, 80 for the sphere–IT plate model.

\[
V_{EDL} = \frac{e_d a_p}{4} \left[ 2\Psi_1 \ln \left( \frac{1 + e^{-kh}}{1 - e^{-kh}} \right) + (\Psi_1^2 + \Psi_2^2) \right] \ln(1 - e^{-2kh})
\]

(6)

\[
V_{EDL} = \frac{e_d a_p}{4} \left[ 2\Psi^2 \ln \left( \frac{1 + e^{-kh}}{1 - e^{-kh}} \right) + (2\Psi^2 \ln(1 - e^{-2kh})) \right]
\]

(7)

\[
V_{EDL} = 2\pi\varepsilon_0\varepsilon_a\sigma_e^2\ln(1 + e^{-h/k})
\]

(8)

Equation 6 is valid for potentials less than 60 mV, when the double-layer thickness is less than the fine particle size, which is true in most scenarios of fine migration in sandstone porous media. $\Psi_1$ and $\Psi_2$ are the surface and compact layer potentials, respectively, and can be replaced by the measured value of the zeta potential ($\zeta$-potential) to develop eq 7. Chequer et al.\textsuperscript{90} presented a new model to calculate $V_{EDL}$ based on the clustered finest sphere–IT plate model, as shown in eq 9.

\[
V_{EDL} = -n160.4 e_m e_0 k_BT \left( \frac{k_BT}{ze} \right)^2 \gamma_p e^{-kh}
\]

(9)

For $V_{EDL}$ calculations based on the kaolinite platelets–IT-plate model, Gregory\textsuperscript{10} presented a different model, as shown in eq 10.

\[
V_{EDL} = \frac{64\eta_0 k_BT}{k} \gamma_p e^{-kh}
\]

(10)

**Born Repulsion ($V_{BR}$) Interaction Energy.** In a colloidal system, when particles approach and are about to contact each other, a short-range repulsive potential called the Born repulsion potential ($V_{BR}$) is generated because their electron clouds overlap. This potential is quite sensitive to the structure of surfaces in contact and permeating liquid. Formulations to quantify the Born repulsion potential for the previously described sphere–IT-plate system have been presented by Ruckenstein et al. and Schumacher et al.\textsuperscript{111,112} in eq 11 and Mahmood et al.\textsuperscript{111} in eq 12.

\[
V_{BR} = \frac{A_H}{7560} \left( \frac{\sigma}{a_p} \right) \left[ \frac{8 + H}{2 + H} + \frac{6 - H}{H^2} \right]
\]

(11)

For clustered fine movement in the porous medium, Chequer et al.\textsuperscript{90} presented a new model to calculate $V_{BR}$, as shown in eq 13.

\[
V_{BR} = -\frac{n\pi a_p^2 \sigma^6}{45h^6}
\]

(13)

A separate model for the natural kaolinite platelets–IT-plate configuration has been presented by Mahmood et al.\textsuperscript{111} as eq 14.

\[
V_{BR} = \frac{A_{Hw} \sigma^6}{360h^6}
\]

(14)

To compute Born repulsion accurately for the fines–rock–fluids system configuration, $A_H$ and $\sigma$ (atomic collision diameter, nm) must be known precisely. An average value used for $\sigma$ in the calculation of $V_{BR}$ is around 0.5 nm. Generally, $V_{BR}$ has a very small impact on the $V_T$ and can be neglected in comparison to other electrostatic potentials ($V_{LVW}$ and $V_{EDL}$) if the separation distance is greater than 1 nm.

**RESULTS AND DISCUSSION**

The EDL expands because of alteration in the pH and ionic strength of the solution, which affects the repulsive force. In this section, we study the effect of these parameters on the total force and fine migration. As mentioned before, the application of NPs increases attractive energy by changing the surface potential to control fine migration, which is also investigated, and results are shown in this section using the DLVO approach.

**pH of Solution.** The pH of colloidal dispersions is one of the most important factors that affect the repulsive force, and it is indicated by the change in the zeta potential of the system. Zeta potential is generally positive at low pH values (acidic region), and with increasing pH, it becomes negative because of the presence of excess OH$^-$. There is a specific pH, where the zeta

![Figure 6. Change in zeta potential with pH of the solution (reproduced with permission from Woodhead Publishing, 2016; pp 299–325).](https://dx.doi.org/10.1021/acsomega.0c03943)

| Table 1. Zeta Potential Data Based on pH of the System |
|-----------------------------------------------|
| **ionic strength** | **pH** | **ζ-potential (mV)** | **source** |
| 2 M (8 wt % NaCl + 2 wt % CaCl$_2$) | 2 | −7 | Singh and Mohanty\textsuperscript{115} |
| 3.5 | −11 |
| 5 | −16 |
| 6 | −22.5 |
| 6.5 | −26 |
| 7 | −28 |
| 8.5 | −32 |
potential becomes zero, which is called the point of zero charge (PZC) or the isoelectric point (IEP), as shown in Figure 6. For a pH higher than the PZC, the surface charge becomes negative, which means a repulsive force that leads to the separation of fines. NPs can shift the PZC by changing the surface forces, so they can be used to control the repulsive force and the detachment of fines. The PZC for SiO$_2$ NPs is around pH = 2.5 – 3. For alumina, it lies between 7.5 and 9, and for MgO, it is around 12 – 13. Hence, for highly alkaline conditions, the application of MgO NPs prevents fine detachment even at high pH values.

Table 2. Effect of Ionic Strength on EDL Thickness

| NaCl $I_s$ (M) | $k$ | $\zeta$-potential (mV) |
|---------------|-----|-----------------------|
| 0.6           | 0.4 | $-17.9$               |
| 0.4           | 0.5 | $-21.3$               |
| 0.2           | 0.7 | $-24.3$               |
| 0.1           | 1   | $-30.5$               |
| 0.05          | 1.4 | $-33.7$               |
| 0.025         | 2   | $-34.0$               |
| 0.01          | 3   | $-34.3$               |

Table 3. Zeta Potential Data for Analysis

| salt   | $I_s$ (M) | $\zeta$-potential (mV) | source       |
|--------|-----------|------------------------|--------------|
| NaCl   | 0.3       | $-16$                  | Chequer et al.  |
|        | 0.25      | $-18$                  |              |
|        | 0.2       | $-19$                  |              |
|        | 0.1       | $-23$                  |              |
|        | 0.02      | $-24.3$                | Assel et al.  |
| CaCl$_2$ | 0.02       | $-8.3$                 |              |
| MgCl$_2$ | 0.02       | $-6.09$                |              |

Table 4. Constants

| parameter                   | symbol | value               |
|-----------------------------|--------|---------------------|
| fine particle radius        | $a_u$  | $5 \times 10^{-7}$ m |
| fluid temperature           | $T$    | 297.15 K            |
| pH of solution              | pH     | 7                   |
| Boltzmann constant          | $k_b$  | $1.38 \times 10^{-23}$ J K$^{-1}$ |
| Hamaker constant            | $A_H$  | $3 \times 10^{-21}$ J |
| permittivity of free space  | $\varepsilon_r$ | $8.85 \times 10^{-12}$ C$^2$ J$^{-1}$ m$^{-1}$ |
| electron charge             | $\varepsilon$ | $1.6 \times 10^{-19}$ C |
| Avogadro’s number           | $N_A$  | $6.02 \times 10^{23}$ mol$^{-1}$ |
| pi                          | $\pi$  | 3.1416               |
| atomic collision diameter   | $\sigma$ | 0.5 nm              |
pH Sensitivity Analysis Based on the DLVO Model.

During alkaline flooding EOR, the alkali generates in situ surfactants that reduce the oil–water interfacial tension to maximize oil recovery. However, alkaline flooding in sandstones may cause fine migration problem because of the change in surface potential caused by an alteration in pH. As the pH of the system increases during alkaline flooding, the repulsive force between fine particles and sand grains increases, and the corresponding zeta potential decreases (becomes more negative) because of excess OH$^-$/1 in the system. This mechanism results in fine detachment and migration during alkaline flooding. The changes in zeta potential caused by increasing the pH of a high-salinity system, composed of a 2 M solution of 8 wt % NaCl with 2 wt % CaCl$_2$ and crushed sandstone grains with kaolinite particles, were measured and shown in Table 1. The DLVO model is used to calculate total energy for the case shown in this table and analyze the effect of pH values ranging from 2 to 8.5, as shown in Figure 7. Surface
forces have been quantified, and the DLVO model has been applied to determine a critical pH value above which fine migration may begin.

Table 5. Hamaker Constant Data

| author                   | year | base liquid                  | pH  | T (°C) | system            | $A_H$ (J) | experimental/theoretical |
|--------------------------|------|------------------------------|-----|--------|-------------------|----------|-------------------------|
| El-Monier and Nasr-El-Din | 2011 | distilled water              | 12  | 149    | kaolinite–quartz  | $1.61 \times 10^{-20}$ | experimental          |
| Habibi et al.            | 2012 | water                        | 7   | 25     | glass beads–water | $6 \times 10^{-21}$   | theoretical           |
| El Badawy et al.         | 2012 | 0.03 M NaCl                  | 7   | 25     | metallic NPs      | $6.04 \times 10^{-20}$| experimental           |
| Arab and Pourahbary       | 2013 | water                        | 7   | 25     | glass beads–water | $6 \times 10^{-21}$   | theoretical           |
| Arab et al.              | 2013 | 0.03 M NaCl                  | 6.9 | 25     | glass beads–brine | $1 \times 10^{-21}$   | theoretical           |
| Arab and Pourahbary       | 2014 | water                        | 7   | 25     | glass beads–water | $6 \times 10^{-21}$   | theoretical           |
| Habibi et al.            | 2014 | 0.03 M NaCl                  | 6.5–7| 25     | sand–NaCl         | $1 \times 10^{-20}$   | theoretical           |
| Xie et al.               | 2014 | 0.2 wt % NaCl                | 8.1 | 65     | oil/silica in water | $8 \times 10^{-21}$  | theoretical           |
| Yang et al.              | 2016 | water                        | 7   | 25     | kaolinite and quartz | $2 \times 10^{-20}$ | theoretical           |
| Mahani et al.            | 2017 | 0.3 M NaCl                   | 7   | 25     | glass beads–brine | $1 \times 10^{-20}$   | theoretical           |
| Hasanajee et al.         | 2017 | 0.355 M diluted sea water    | 7   | 25     | limestone–brine   | $1 \times 10^{-19}$   | theoretical           |
| Xie et al.               | 2018 | NaCl, MgCl$_2$, and CaCl$_2$ | 4–10| 25     | shale–oil         | $0.81 \times 10^{-20}$| theoretical           |
| Huang et al.             | 2018 | 2 wt % KCl                  | 7   | 25     | coal–brine        | $4.62 \times 10^{-20}$| theoretical           |
| Sanaei et al.            | 2019 | 0.01 M NaCl                  | 7   | 25     | carbonate–brine   | $1.3 \times 10^{-20}$ | theoretical           |
| Chequer et al.           | 2019 | 0.6 M NaCl                   | 7   | 25     | kaolinite–sand    | $1.49 \times 10^{-20}$| experimental          |
| Takeya et al.            | 2020 | 0.1 M ALSW                   | 7   | 25     | calcite–brine     | $6.6 \times 10^{-21}$ | theoretical           |
| Tangpartkul et al.       | 2020 | 0.0005 M NaCl brine          | 7   | 25     | clay–sand         | $2 \times 10^{-21}$   | theoretical           |
| Gomez-Flores et al.      | 2020 | 0.001 M NaCl                 | 7   | 25     | silica–brine      | $3.91 \times 10^{-21}$| theoretical           |
| Peng et al.              | 2020 | 0.007 M SDS and 0.0005 M NaCl| 7   | 25     | surfactant–water  | $5.2 \times 10^{-20}$ | experimental           |

The critical pH value to initiate fine migration is between 6 and 6.5 for this case. Thus, maintaining the system pH below 6 can prevent fine migration. This calculation shows that the application of alkaline flooding leads to fine migration because in alkaline flooding the pH will be more than 7.

**Ionic Strength ($I_s$).** The ionic strength of the solution affects the expansion and thickness of the EDL, which further affects the repulsive force and total energy of the system. The higher the ionic strength of the solution, the more compressed the EDL becomes. For the quantification of EDL repulsion, the ionic strength and the corresponding experimental zeta potential data have been taken from research work by Chequer et al. The effect of decreasing the ionic strength on the Debye length ($k$), which is an indicator of EDL thickness, is shown in Table 2. As the solution salinity decreases, EDLs around the fine particles and the sand grains both expand simultaneously, generating...
more repulsion between the fines and the sand grains. Consequently, detachment of the fines occurs.

The low zeta potential confirms the high repulsive force in these conditions. Figure 8 demonstrates the increase in repulsive force when the ionic strength is reduced from a high-salinity (0.6 M) to low-salinity environment (0.01 M). However, a decrease in brine salinity does not affect van der Waals attraction at any molarity, as depicted by the DLVO model in Figure 9a. Both energy contributions result in the total energy for the system incorporating constant attraction and variable repulsion at different salinities, indicating a critical salinity from 0.2 to 0.4 M, below which we may have fine detachment, as shown in Figure 9b.

For systems containing silica glass beads, fines, and 0.02 M NaCl, CaCl₂, and MgCl₂ solutions, zeta potentials have been measured by Assef et al. Data for 0.3–0.1 M NaCl have been measured by Chequer et al., as shown in Table 3. We applied the DLVO approach to estimate the CSC value for these cases. Calculations of attractive and repulsive forces have been made for salinities from 0.3 to 0.02 M, and the results are shown in terms of the dimensionless V₉ of the system. Data used in the model are given in Table 4.

Table 7. Zeta Potential Data in the Presence of NPs

| authors | year | NP type | NP size (nm) | NP conc. | base liquid | pH | T(°C) | environment | ζ-potential (mV) | apparatus/method |
|---------|------|---------|--------------|----------|-------------|----|-------|-------------|----------------|-----------------|
| Rouxel et al.132 | 2011 | α-Al₂O₃ | 13 | 0.05 wt % | DI water | 7.2 | 25 | water | −34 | Zetasizer |
| Priya et al.138 | 2012 | CuO | 40–60 | 0.016 vol % | DI water | 12 | 25 | water | −30 | Zetasizer |
| El Badawy et al.122 | 2012 | H₂−Ag | 13 | 3 vol % | 0.03 M NaCl | 7 | saline | −22 | Zetasizer |
| Suganthi and Rajan144 | 2012 | ZnO | 30−45 | 1 vol % | DI water | −25 | 25 | water | −49.9 | Zetasizer |
| Habibi et al.71 | 2012 | MgO | 63 | − | DI water | 7 | 40 | glass beads | −5.68 | Zetasizer |
| Ahmadi et al.69 | 2013 | MgO | 63 | 0.1 wt % | 0.3 M NaCl | 7 | 25 | Berea sand | −7 | Zetasizer |
| SiO₂ | 48 | 0.1 wt % | DI water | 7 | 25 | glass beads | −34 | 12.8 | Zetasizer |
| Al₂O₃ | 43 | 0.1 wt % | DI water | 7 | 25 | glass beads | −24.4 | −11 | Zetasizer |
| Arab and Pourafshary65 | 2013 | ZnO | 30 | 0.03 wt % | DI water | 7 | 25 | glass beads | −44 | 1.57 | Zetasizer |
| Arab et al.67 | 2014 | MgO | 20 | 0.03 wt % | 0.03 M NaCl | 6.9 | 25 | glass beads | −27.6 | −5.7 | Zetasizer |
| Assef et al.66 | 2014 | MgO | 20 | 0.03 wt % | DI water | 7 | 25 | glass beads | −34 | −1 | Zetasizer |
| Bayat et al.155 | 2015 | Al₂O₃ | 40 | 0.005 wt % | DI water | 6.4 | 26 | water | −19.1 | ζ analyzer |
| TiO₂ | 30 | 0.05 wt % | DI water | 6.4 | 26 | water | −9.1 | ζ analyzer |
| SiO₂ | 20 | 0.05 wt % | DI water | 6.4 | 26 | water | −28.1 | ζ analyzer |
| Alomair et al.64 | 2015 | TiO₂ | 50 | 0.1 wt % | 30000 ppm brine | 5.9 | 40 | Berea sand | −13.2 | Zetasizer |
| SiO₂ | 15 | 0.1 wt % | 30000 ppm brine | 5.9 | 40 | Berea sand | −28.3 | Zetasizer |
| Al₂O₃ | 40 | 0.1 wt % | 30000 ppm brine | 5.9 | 40 | Berea sand | −25.3 | Zetasizer |
| NiO | 50 | 0.1 wt % | 30000 ppm brine | 5.9 | 40 | Berea sand | −23.4 | Zetasizer |
| Sabiha et al.136 | 2016 | SWCNT | 2500 L | 0.1 vol % | DI water | −25 | 25 | water | −53.1 | Zetasizer |
| Adil et al.137 | 2016 | ZnO | 55 | 0.1 wt % | 30000 ppm NaCl | 9 | 95 | saline | −20 | Zetasizer |
| Hasannejad et al.77 | 2017 | SiO₂ | 145 | 0.1 wt % | 0.3 M NaCl | 7 | 25 | glass beads | −28 | −25 | Zetasizer |
| Lee et al.158 | 2017 | SiO₂ | 37 | 6.72 wt % | 4.5 M NaCl/NaCl₂ | 8 | 25 | saline | −24 | Zetasizer |
| Al-Ansari et al.117 | 2017 | SiO₂ | 10 | 0.1 wt % | 1 wt % NaCl | 6.3 | − | saline | −40 | −5 | Zetasizer |
| Abdelfatah et al.139 | 2017 | SiO₂ | 10 | − | − | 6 | 25 | − | −22 | − |
| Skoglund et al.140 | 2017 | Ag NPs | 9 | − | DI water | −25 | water | −44 | Zetasizer |
| Choudhary et al.1,141 | 2017 | γ-Al₂O₃ | 20 | 0.1 wt % | DI water | 7.6 | − | water | −36.7 | Zetasizer |
| Upendar et al.142 | 2018 | α-Fe₂O₃ | 20 | 0.05 wt % | 0.001 M NaCl | 6.5 | 25 | saline | −9.7 | EFM |
| Kuang et al.143 | 2018 | Al₂O₃ | 50 | 0.1 wt % | 0.001 M NaCl | 7 | 25 | saline | −39 | ζ analyzer |
| Mansouri et al.130 | 2019 | SiO₂ | 15 | 0.1 wt % | 0.03 M NaCl | 5.9 | 25 | glass beads | −25 | −7.5 | Zetasizer |
| Ma et al.144 | 2019 | SiO₂-g-SPMA | 100 | 0.5 wt % | 5.4 M NaCl + NaCl₂ | 11 | 170 | saline | −54 | Zetasizer |
| Siddiqi et al.62,145 | 2019 | Cu–Al₂O₃ | 270 | 0.01 wt % | DI water | 7 | 23 | water | −48.15 | ζ analyzer |
| Aramendiz and Imqam146 | 2019 | SiO₂ | 20 | 0.75 wt % | 9.5 | 25 | water base mud | −34.66 | ζ analyzer |
| Kumar et al. | 2020 | SiO₂–TiO₂ | 15 | 0.4, 0.05 wt % | DI water | 7 | 90 | water | −45.4 | −34.1 | Zetasizer |
| Wang et al.147 | 2020 | CNCs | 70 | 0.25 wt % | 0.175 M NaCl | 7 | 21 | saline | −60 | Zetasizer |

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Figure 10 shows the effect of changing the salinity from 0.3 to 0.02 M NaCl solutions containing silica glass beads and fine particles to mimic a sandstone formation. The total DLVO energy for each scenario has been calculated using the sphere−IT plate DLVO model. As shown in Figure 10, total interaction energy is attractive for the high-salinity case because of the weak EDL repulsion. As the salinity is gradually decreased from 0.3 to 0.2 M, total system energy shifts from negative to positive, indicating that the EDLs have expanded and overlapped, causing an increase in repulsive forces (which dominated the attractive forces) under low-salinity conditions. It is observed from the DLVO model calculation that the CSC for the NaCl system is in the range of 0.2−0.25 M, which is in close agreement with the experimental value for the NaCl solution.12,60

Nanoparticles. Data concerning various NPs used in different studies, which were performed for fine fixation, wettability modification, interfacial tension (IFT) reduction, drilling mud preparation, and stable NFs formulation, have been collected, and the results have been presented in Figure 11.

It is observed that the maximum number of studies are performed using SiO2 NPs because of their high stability, low toxicity, resistance to pH changes, and easy availability.116 MgO and Al2O3 NPs are next on the list and have been utilized in several studies to mitigate fine release, the migration issue, and IFT reduction, respectively. NPs are used to overcome the fine migration problem in sandstone reservoirs by changing the surface forces, which can be observed by an alteration in zeta potential values before and after the adsorption of NPs onto rock/grain surfaces. Figure 12 presents zeta potential values of glass beads before and after the application of different NPs in low-salinity conditions.60,63,65,67,69,71,77,117 Figure 12 shows that MgO NPs are the best at changing the surface energy and increasing zeta potential. In a few research studies, ZnO, TiO2, and NiO NPs, and combinations of more than one NP, have been used in the form of NFs. Future research will benefit most

Table 8. Zeta Potential Data without NPs

| authors               | year | base liquid          | salinity | pH | T (°C) | environment | ζ (mV) | apparatus/method |
|-----------------------|------|----------------------|----------|----|--------|-------------|--------|------------------|
| Yousef et al.148      | 2012 | seawater             | 600 ppm  | 7−8| 60     | carbonate   | −13    | Zetasizer        |
| Nasralla and Nasr-El-Din 149 | 2012 | NaCl brine           | 0.2 wt % | 7.7 | 25     | Berea SS    | −35    | phase-analysis light-scattering PALS |
| Hussain et al.36      | 2013 | NaCl brine           | 0.5 M    | –  | 25     | sand        | −20    | –                |
| Chen et al.149        | 2014 | formation water      | 14000 ppm| 8  | 25     | limestone   | −15    | phase-analysis light-scattering PALS |
| Xie et al.25          | 2014 | 0.2 wt % NaCl        | 0.2 wt % | 8.1 | 65     | sandstone   | −23.7  | Zetasizer        |
| Xie et al.118         | 2016 | NaCl brine           | 0.2 wt % | –  | 25     | sandstone   | −33    | –                |
| Yao et al.150         | 2016 | distilled water       | 0        | 9.2 | 25     | quartz      | −23    | Zetasizer        |
| Mahani et al.21       | 2017 | diluted seawater      | 0.035 M  | 7  | 25     | limestone   | −11    | Zetasizer        |
| Huang et al.21        | 2018 | KCl                   | 2 wt %   | 7  | 25     | coal−brine  | −12.61 | micro-electrophoresis |
| Sanaei et al.24       | 2019 | NaCl brine            | 0.01 M   | 7  | 25     | carbonate   | −50    | –                |
| Chequer et al.90      | 2019 | NaCl brine            | 0.6 M    | 7  | 25     | sand        | −20    | Zetasizer        |
| Takeya et al.20       | 2019 | NaCl brine            | 0.7 M    | 7.2| 50     | crude oil   | −23    | ζ analyzer       |
| Alghamdi et al.151    | 2020 | smart water           | 5761 ppm | 7.4| –      | carbonate   | −8     | Smoluchowski equation |
| Ruan et al.152        | 2020 | 0.1 M KCl             | 0.1 M    | 7  | 30     | clay−brine  | 4.9    | Zetasizer        |
| Takeya et al.23       | 2020 | ALSW                  | 0.1 M    | 7  | 25     | calcite−brine | −3.72  | Zetasizer        |
| Tangaritkul et al.35   | 2020 | NaCl brine            | 0.000513 M| – | –      | clay−sand   | −50    | Zetasizer        |
| Gomez-Flores et al.92 | 2020 | NaCl brine            | 0.001 M  | 7  | 25     | silica−brine | −39    | –                |
| Peng et al.125        | 2020 | 7 mM SDS and 0.5 m NaCl brine | – | –  | 25     | surfactant−water | −80    | –                |

Figure 17. Zeta potential correlations for sandstone.

Figure 18. Close agreement between measured and calculated zeta potentials.

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from using different types of single and hybrid NFs. In an experimental study, Assef et al.66 used MgO NPs to mitigate the adverse effects of fine migration that were noticed under low-salinity flow conditions (0.02 M NaCl).

They observed experimentally that the application of only 0.0075 wt % MgO NPs retained around 97% of in situ fine particles even in highly alkaline conditions of pH = 9.2. Retention of fine particles was attributed to increasing attractive forces between fines and glass beads in the presence of MgO NPs. To validate the experimental results, we applied the DLVO theory to quantify the total energy of the system, which is presented in Figure 13. Surface force quantification and analysis showed that the application of NPs increased the attractive forces between fines and grains and consequently the $V_T$ of the system decreased, indicating a shift from repulsion to attraction.

**Divalent Ions.** The presence of divalent ions (Ca$^{2+}$, Mg$^{2+}$) in the solution is beneficial and can suppress the EDL thickness, resulting in reduced repulsive force as compared to monovalent ions (Na$^+$) for the same salinity. Xie et al.118 performed experimental research and measured zeta potentials for a solution containing crushed sand particles in the presence of monovalent (Na$^+$) and divalent ions (Ca$^{2+}$, Mg$^{2+}$) separately. They observed that divalent ions suppressed the EDL which was formed around the sand grain and hence resulted in reduced repulsion, which is indicated by low zeta potential (absolute value), as shown in Figure 14.

Similar results for zeta potential data were observed in the studies performed by Shehata and Nasr-El-Din119 and Gulgonul120 on sandstone and natural hydrophobic Teflon, respectively, in the presence of monovalent and divalent ions (Ca$^{2+}$, Mg$^{2+}$). Assef et al.66 also utilized CaCl$_2$ and MgCl$_2$ solutions and performed similar experiments. Quantification of surface forces has been done, and the DLVO model has been applied. Figure 15 depicts the relative comparison of $V_T$ for monovalent and divalent ions in the electrolyte solution. The presence of divalent ions reduces the repulsive force and suppresses the expansion of the EDL even under low-salinity conditions because of their high valence charges. Divalent ions in the solution can help to design a lower value of the CSC, which means that low-salinity flooding benefits can be achieved without fine migration in sandstone reservoirs by tuning the composition of the injection brine.

**Interactive Parameters for the FBR System.** Van der Waals particle–particle attraction is produced when particles/molecules come close to each other in a medium. The Hamaker constant is a coefficient that relates this interactive energy among particles, whereas zeta potential ($\zeta$) is a measure of the surface charge of the particles and an indicator of increase and decrease of the repulsive force. The following section discusses these parameters in detail.

**Hamaker Constant ($A_H$).** Hamaker constant is dependent on the integrated system of the fine particle, its shape, pore surface, aqueous medium type, and salinity, and crude oil properties.80 The Hamaker constant is determined experimentally with great caution based on the specific system configuration. The typical values for $A_H$ are found in the range of $10^{-21}$ to $10^{-19}$ J. These experimental values are in close agreement with the theoretical calculations of Israelachvili.80,121

Equation 15 is used to calculate this constant using experimental data.

$$A_H = \frac{3}{4} k_B T \left( \frac{e_1 - e_3}{e_1 + e_3} \right) \left( \frac{e_2 - e_3}{e_2 + e_3} \right) \frac{3h\nu_c}{8\sqrt{2}} \left( \eta_1^2 - \eta_3^2 \right)$$

where $e_i$ is the static dielectric constant, $\eta$ is a refractive index, and $h\nu_c$ is the Planck constant. $\eta_1$ is the constant value of electronic adsorption frequency equal to $3 \times 10^{-15}$ s$^{-1}$. Table 5 presents studies where Hamaker constants were calculated and a few studies where experimental approaches have been used to measure $A_H$ based on the system’s actual configuration.80,93,122,123 A sensitivity analysis for $A_H$ has been performed and total interaction energy has been quantified in Figure 16. A high Hamaker constant leads to more attraction, whereas its low value is related to reduced attraction, as demonstrated by the DLVO model in Figure 16a. Also, particle deposition onto a rock’s surface is affected by the Hamaker constant of the interactive system. Elimelech et al.124 found that at low salinity, the range of EDL is much higher than that of van der Waals attraction, and the rate of particle deposition is mainly controlled by double-layer repulsive energy.

However, at high-salinity, van der Waals attractive forces are more effective, and as a result, the rate of particle deposition is controlled by $A_H$. Figure 16b illustrates the effect of different $A_H$ values on particle deposition at different salinities. More particle deposition is observed because the repulsion is reduced at high $A_H$ in a high-salinity environment. Hence, it is recommended that the experimental value of Hamaker constant be used in modeling, for the fine particle, rock grain, temperature, and an aqueous medium that could be distilled water or brine of specific salinity, to get accurate results using the DLVO model.

**Zeta Potential ($\zeta$).** Zeta potential is an important indicator of surface charge and is used for the quantification of the electrostatic repulsive energy between dispersed fines and sand grains. It is the potential difference between the surfaces because of the electrical difference between a particle surface and points away from the particle in the fluid at the boundary of the slipping plane,126,127 as shown in Figure 5. This potential is also known as electro-kinetic potential in colloidal dispersions. It is worth mentioning here that the measured zeta potential of clay particles (fines) is negative and is an important parameter that provides information about the charge on the particle surface, colloidal system stability in an ionic environment, electrostatic forces between particles and the rock surface, and interaction energy between NPs and formation fines in porous media. Zeta potential is an input parameter in the calculation of the EDL repulsive force and must be determined experimentally with great accuracy. It is usually measured with a Zetasizer, which uses the electrophoretic mobility (EFM) concept based on the Helmholtz Smoluchowski equation (shown in eq 16).

$$\zeta = \frac{U_z 3\eta}{2eF\kappa a}$$

Tables 7 and 8 present the zeta potential data collected from several previously published research articles that show the widespread use of this parameter. It is evident from the data that in all the studies, the zeta potential of the system has been determined with the help of the Zetasizer/zeta potential analyzer, which is a high-technology and expensive apparatus.
being used worldwide. It measures the electrophoretic mobility and automatically converts it to provide the direct zeta potential value of the system under study. No comprehensive correlation is available to directly calculate zeta potential as a function of system configuration (sand, glass beads, fines, kaolinite, calcite, etc.), salinity, viscosity, pH, and temperature.

Based on the data available in Table 8 for similar neutral systems (pH ≃ 7–8.5) containing sandstone and NaCl solutions of different $I'_{Na}$ two correlations are developed to estimate zeta potential for high-salinity and low-salinity conditions, as shown in Figure 17. Equations 17 and 18 present the developed correlations for low-salinity (0–0.9 M) and high-salinity (0.1–0.9 M approx.) conditions, respectively.

\[
\zeta\text{-potential (mV)} = 96.315 \times M - 38.786 + \text{error} \quad (17)
\]

\[
\zeta\text{-potential (mV)} = 25.658 \times M - 34.053 + \text{error} \quad (18)
\]

These correlations are validated for specific salinity range with laboratory data taken from different studies with similar conditions (sandstone, pH ≃ 7–8.5, 25 °C). and the results are shown in Table 6. Model results are in close agreement with the laboratory measured zeta potential, with less than 10% error for most of the data points, as shown in Figure 18.

Hence, the generated models can be used to estimate the zeta potential of similar systems, eliminating the need to perform extensive experiments. The calculated zeta potential of sand grains demonstrates the contraction and expansion of the EDLs around fines and sand grains under different salinity conditions. Regarding the practical viewpoint, the developed correlations can considerably benefit the reservoir engineers and production chemists who are involved in the modeling and designing of injection water chemistry/recipe and operation in sandstone reservoirs to avoid fine migration while maintaining designed injectivity.

**CONCLUSIONS**

The DLVO modeling technique based on the quantification and analysis of surface forces is a powerful tool that helps in the analysis of fine migration and control in sandstone reservoirs during low-salinity water injection and alkaline flooding without extensive experimentation. The results can be summarized as follows:

1. Based on surface force analysis of the FBR system for the solution containing monovalent and divalent ions, this tool can predict the CSC for the injection fluid below which the total interaction energy for the system becomes repulsive and the fine migration starts within the porous media.
2. Attractive and repulsive forces are affected by fine particle size, ionic strength, types of ions, pH, and the viscosity of the flowing fluid, and all these factors must be considered for the quantification of surface forces.
3. The application of NPs changes the surface energy and is a promising technique to control and optimize the fine migration with reduced critical salinity. Surface force analysis shows an increase in attractive interaction energy after the application of NPs.
4. Zeta potential of the FBR system and Hamaker constant are important indicators for any change in attractive and repulsive forces. Precise measurements of these influential parameters can reduce uncertainty in the results and help to provide a better outcome.

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**Notes**

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**NOMENCLATURE**

- $A_H$, Hamaker constant, J
- $a_p$, particle radius, nm
- $C$, concentration of ion, M
- $e$, electron charge, C = 1.60 × 10⁻¹⁹ C
- $F(k_c)$, Henry’s function
- $F(g)$, gravitational force, kg·m·s⁻²
- $F_e$, net electrostatic force, kg·m·s⁻²
- $h$, separation distance, nm
- $H$, dimensionless distance = $h/a_p$
- $I_d$, ionic strength of permeating fluid, $mV$
- $k_B$, Boltzman constant, J·K⁻¹
- $k$, inverse Debye length, m⁻¹ = $\sqrt{\frac{e_k T}{2N_e e_i^2}}$
- $n$, number of fines in a cluster
- $N_A$, Avogadro’s number = 6.02214 × 10²³ mol⁻¹
- $R$, average pore throat radius, m
- $T$, absolute temperature, K
- $U_e$, electrophoretic mobility, V·m⁻¹
- $V_{LVW}$, London–van der Waals potential
- $V_{EDL}$, electrical double-layer potential
- $V_{Bd}$, Born repulsion potential
- $\nu_e$, electronic adsorption frequency, s⁻¹
- $z$, valence of ion
- $v$, flowing fluid velocity, m/s
- $\epsilon$, dielectric constant in J·m⁻³
- $\psi$, potential, mV
- $\eta$, refractive index
- $\zeta$, zeta potential, mV
- $\sigma$, atomic collision diameter, nm
- $\epsilon_{so}$, relative static permittivity of water
- $\epsilon_{v}$, permittivity of the vacuum
- $\eta$, viscosity of solution, kg·m⁻¹·s⁻¹
- $\gamma_v$, tanh($\zeta z / 4k_BT$)
- $\gamma_v^*$, tanh($\zeta z / 4k_BT$)

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