

MOBILIZATION OF QUARTZ FINES IN POROUS MEDIA¹

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Abstract—The onset of the mobilization of fine particles of quartz (fines) in sandpicks was determined by comparing the theoretically calculated hydrodynamic and colloidal forces acting on a fines particle near a representative sand grain. The results show that the mobilization of fines depends strongly on the chemistry of fluids present in the reservoir. Specifically, a critical electrolyte concentration exists for mobilization, which depends on the pH. For large particles of fines and relatively high fluid velocity, the mobilization of fines may depend on the fluid velocity, but in a narrow range of electrolyte concentration. The types of interactions between the fines and sand grain surfaces were corroborated by direct visual observations using a traveling microcell pack.

The mobilization of fines in sandstones leads to a reduction of permeability (i.e., a reduction of the hydraulic conductivity).

Key Words—Colloidal forces, Fines particles, Electrolyte concentration, Hydrodynamic forces, Mobilization, Porous media, Quartz.

INTRODUCTION

The mobilization of fine-grained particles (hereafter referred as “fines” or quartz fines) and their subsequent entrainment and redeposition in porous media can lead to a substantial decline in the productivity of producing petroleum reservoirs. Similarly, soil permeability can be reduced and permeability at some freshwater-seawater interfaces may be reduced due to the changes in the chemistry of the fluids (Goldenberg *et al.*, 1984; Goldenberg, 1985; Goldenberg and Margaritz, 1983). In petroleum reservoirs, these fine-grained particles are commonly small particles of quartz and/or clays which may become entrained in the flowing (displacing) fluid and transported through the porous formation. Most of these particles, however, may not reach the producing well, but become trapped at pore constrictions, thereby causing severe plugging of the pores and reducing the permeability of the formations (Jones, 1964; Krueger *et al.*, 1967). For example, the sensitivity of water in sandstone is a phenomenon in which the permeability of argillaceous sandstones decreases sharply when freshwater replaces the salt water initially present in the formations (Mungan, 1965, 1968; Khilar and Fogler, 1981). This phenomenon is similar to that found at the contact between freshwater and seawater in aquifers (Goldenberg *et al.*, 1984); however, the reduction in permeability here may be attributed to either clay swelling, clay particle migration, or a combination of both effects.

Significant concentrations of fines can reduce the permeability of petroleum reservoirs. In addition, alkali flooding and steam injection can increase the dissolved silica concentrations and the subsequent formation of colloidal silica (Reed, 1980).

In the present work, the hydrodynamic and colloidal

forces between quartz fines and sand grain surfaces were analyzed to determine the criteria for mobilization of fines using Happel's model (1958) and the Derjaguin-Landau and Verwey-Overbeek (DLVO) theory. Presumably, the fines are initially attached to sand grain surfaces by van der Waals attractive forces, which predominate at high electrolyte concentrations. This assumption is reasonable, inasmuch as the salinity of most petroleum reservoirs is relatively high. Also, Goldenberg (1985) and Goldenberg *et al.* (1984) found sharp decreases in hydraulic conductivity (or an equivalent reduction in permeability) if aquifer water of very low salinity (10^{-3} M NaCl) replaced the seawater (salinity ~ 0.5 M NaCl) initially present in the formation. The effects of pH and fluid velocity on the critical electrolyte concentration for the mobilization of fines were also investigated. To verify the theoretical analysis, direct visual observations were made using a traveling microcell pack invaded by suspensions of quartz fines at fixed electrolyte concentration and pH.

THEORETICAL BACKGROUND

Model of flow in porous media for calculating hydrodynamic forces

If flow takes place in a porous medium, a fines particle attached to the surface of a sand grain is subjected to hydrodynamic forces. As a consequence, a fines particle very near a sand grain will be subjected to a shear force due to uniform shear flow, as given by Goren and O'Neill (1971) and Spielman and Fitzpatrick (1973):

$$F_{SH} = 6\pi\mu a_p^2 \cdot \frac{3 A_s U \sin \theta_p}{2 a_s} \cdot (H + 1)F(H), \quad (1)$$

where μ is the viscosity, a_p is the radius of the fines particle, A_s is a dimensionless porosity parameter (Happel, 1958), U is the superficial velocity of the fluid, θ_p is the angle between the radial coordinate of the center of the fines particle and the direction of the incoming flow, $F(H)$ is a universal hydrodynamic function of the dimensionless gap, $H = h/a_p$ (where h is the separation distance between a fines particle and the sand grain surface), and a_s is the radius of the sand grain.

This force is a tangential force with respect to the sand grain surface (see Figure 1). Visser (1980) and Hubbe (1985) recognized that the first stage in the detachment of colloidal particles from solid substrates under laminar conditions involves a rolling movement caused by the fluid flow. The removal or detachment of a fines particle due to the fluid flow invading the porous medium is mainly dictated by this force. Other factors may contribute to particle detachment, such as surface roughness and local fluid flow disturbances that may induce lifting forces (Hubbe, 1985) (not considered in the present work). Consequently, in the following analysis, the force F_{SH} is compared with the colloidal forces between fines particles and sand grain surfaces in order to determine the critical conditions for the mobilization of fines.

Colloidal forces

The behavior of a fines particle very near a sand grain surface is a function of colloidal forces (double layer repulsive forces and van der Waals attractive forces) and the hydrodynamic forces outlined above. As will be shown, the colloidal forces depend strongly on the electrolyte concentration. The double layer repulsive force between a spherical fines particle and a sand grain surface (the latter considered a flat-plate if $a_p \ll a_s$) was given by Gregory (1975):

$$F_{DL} = 2\pi a_p \cdot \frac{nkT}{\kappa} \cdot [(y_1^2 + y_2^2) \cdot (1 - 1/\tanh(\kappa h)) + 2y_1 y_2 / \sinh(\kappa h)], \quad (2)$$

where n is the number of ions per unit volume; k is Boltzmann's constant; T is the absolute temperature; y_1 and y_2 are the reduced surface potentials of the sand grain and fines particle, respectively ($y_i = ze\psi_i/kT$, where z is the valency of the counterions, e is the electron charge, and ψ_i is the surface potential); and κ is the Debye-Huckel reciprocal length parameter, $\kappa^2 = 2e^2nz^2/\epsilon kT$ (where ϵ is the permittivity of the fluid medium).

The van der Waals force between a fines particle and a sand grain surface can be expressed as follows (Gregory, 1981):

$$F_{VW} = -2\pi a_p \cdot \frac{A}{12\pi h^2} \cdot \frac{1}{(5.32h/\lambda + 1)}, \quad (3)$$

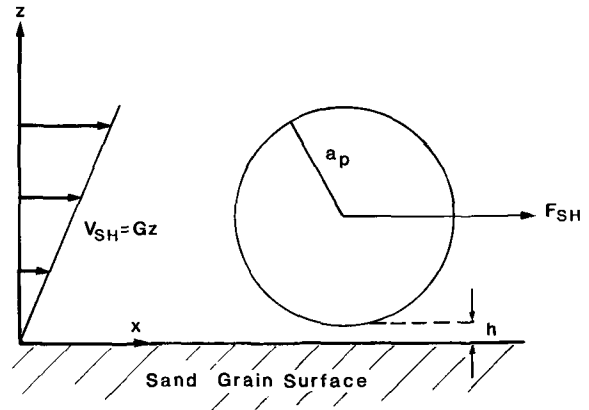


Figure 1. Schematic representation of the hydrodynamic force acting on a spherical particle near the sand grain surface due to the shear flow depicted. Here, fluid velocity is given by $V_{SH} = Gz$, where G is the shear rate.

where λ is the London wavelength, which is ~ 100 nm (Gregory, 1981). In the calculations, the value $A = 1.7 \times 10^{-20}$ J has been used for the Hamaker constant (Hough and White, 1980). Born repulsion force, which is an extremely short-range interaction ($1-10$ Å), has not been included in this work because it has no significant effect in the analysis. The net colloidal force between a fines particle and a sand grain surface is the superposition of the above two forces, namely:

$$F_T = F_{DL} + F_{VW}. \quad (4)$$

F_{DL} can be evaluated with the assumption that the surface potentials can be approximated by the zeta potentials (ζ), which give a good indication of the magnitude of the repulsive interaction force between particles (Hunter, 1981). The zeta potentials for fines can be obtained from electrophoretic measurements and for a sand grain from streaming potential measurements. The zeta potentials were obtained as described below.

EXPERIMENTAL

The zeta potential of the sand grain (Fisher silica sand S-150) was obtained by measuring the alternating streaming potential generated by a laminar sinusoidal flow through an acrylic packed cell 1.2 cm in diameter and 5 cm long. In this technique, the electrodes' polarization was reduced, because an alternating current was produced. In this manner the absolute zeta potential was calculated using the expression (Sears and Groves, 1978):

$$E_s = \frac{\Delta P \epsilon K \sigma \rho \zeta^2}{\mu}, \quad (5)$$

where E_s is the streaming potential, ΔP is the pressure

Table 1. Zeta potentials, ξ (in mV), for a porous silica medium.¹

NaCl concentration (M)	pH 3	pH 7	pH 9
10^{-1}	7	-15	-20
10^{-2}	10	-32	-47
10^{-3}	16	-65	-80

² Fisher silica sand grade S-150, grain size $\sim 180 \mu\text{m}$.

drop across the porous plug, σ is a complicated function of frequency, K is the dielectric constant of the fluid, and ρ is the resistivity of the porous plug. The zeta potentials of the sandpack obtained using this technique are given in Table 1. These data agree quite well with data reported by Gaudin and Fuerstenau (1955).

Fine particle-size quartz was obtained by screening Fisher silica sand (S-152) through 400 mesh to get the $<36\text{-}\mu\text{m}$ fraction. The $<36\text{-}\mu\text{m}$ size material was then settled in distilled water and the $<1\text{-}\mu\text{m}$ size fraction collected. The zeta potentials of $<1\text{-}\mu\text{m}$ material were obtained from the measured electrophoretic mobilities, U_E , using a microelectrophoresis apparatus (Rank Brothers Mark II) equipped with a rectangular cell. Samples were prepared at desired electrolyte (NaCl) concentrations, and the pH was adjusted before each measurement. The zeta potentials were calculated using the Smoluchowski equation, $U_E = \epsilon\xi/\mu$ (symbols defined above). The results are given in Table 2. The values are different from those in Table 1, because large grains probably did not expose a surface chemically equivalent to that of the fines. Furthermore, the roughness of the sand grain surfaces may also have affected the zeta potentials. As shown in the two tables, the zeta potential varied significantly with both electrolyte concentration and pH. This behavior is important inasmuch as it will determine the structure of the double layer between the solid and liquid and eventually provided information on the type of interaction between the fines and sand grain surfaces.

Visualization experiments on the movement of fines in porous media were carried out using the traveling microcell apparatus. The microcell was constructed by placing sand in a small chamber consisting of a glass slide $5 \times 7.5 \text{ cm}$ and a thin cover glass $2 \times 5 \text{ cm}$. The thickness of the packed bed was $\sim 0.15 \text{ cm}$. The microcell was then mounted on the vertical traveling stage, and the sandpack was saturated with a solution of the same electrolyte concentration and pH as that of the fines suspension that was to be injected afterwards. A syringe pump (model 341A, Sage Instruments) was used to inject the fines suspension both from the top and from the bottom of the cell. Fines movement and deposition in the porous medium were video recorded, and photographs were taken using a Polaroid camera at 250 and $320\times$ magnifications.

Table 2. Zeta potential, ζ (in mV), for fine-grained quartz particles.¹

NaCl concentration (M)	pH 3	pH 7	pH 9
10^{-1}	-12	-35	-42
10^{-2}	-16	-60	-69
10^{-3}	-24	-78	-87

¹ Particle size $\sim 1 \mu\text{m}$.

RESULTS AND DISCUSSION

Hydrodynamic forces

The mobilization of fines depends on the closest separation distance (h_0) between fines and sand grain surfaces below which colloidal forces become infinitely repulsive (Frens, 1978). As a consequence, a fines particle cannot approach the grain surface at distances $h < h_0$. Considering the surface roughness of the grains (and fines as well), the magnitude of h_0 was probably in the range of 20 \AA . Such relatively large closest-separation-distance has been also reported by other investigators (Hubbe, 1985; Gotoh *et al.*, 1983).

The hydrodynamic forces acting on the quartz fines very near the sand grain surface were evaluated to determine the conditions for detachment. Here the fine-grained particles were considered to be spherical particles, which is shown by Eq. (1). Computations for porosities $\phi = 0.20$ to 0.40 , a superficial velocity of 1 cm/hr , a fines particle size of $1 \mu\text{m}$, and grain diameters of the porous media of 50 to $200 \mu\text{m}$ showed that F_{SH} ranged from 0 to 2 pN per particle. For larger fines particles, for example, assuming a typical fluid velocity near a well bore of 20 cm/hr and a sand grain diameter of $100 \mu\text{m}$, F_{SH} was in the range 40 to 80 pN for $a_p = 5 \mu\text{m}$ and $\phi = 0.30$. Assuming a high velocity of 200 cm/hr (during steam injection, for example), the hydrodynamic forces were of the same magnitude as the colloidal forces (see below). The mobilization of fines depended on fluid velocity if the superficial fluid velocity was high or if the displacing fluid was a high viscosity oil.

Colloidal forces

The colloidal forces between a fines particle and a sand grain surface were calculated using Eqs. (2) and (3) and the zeta potentials given in Tables 1 and 2. Figure 2 shows the colloidal forces between a fines particle of $a_p = 1 \mu\text{m}$ and a sand grain surface as a function of separation distance h at different pHs (3, 7, and 9) and NaCl concentrations, respectively. As expected the interactions strongly depended on the chemistry of the surrounding fluids (electrolyte concentration and pH). For example, at pH 3.0, the colloidal forces were significantly attractive regardless of the electrolyte concentration, because the zeta poten-

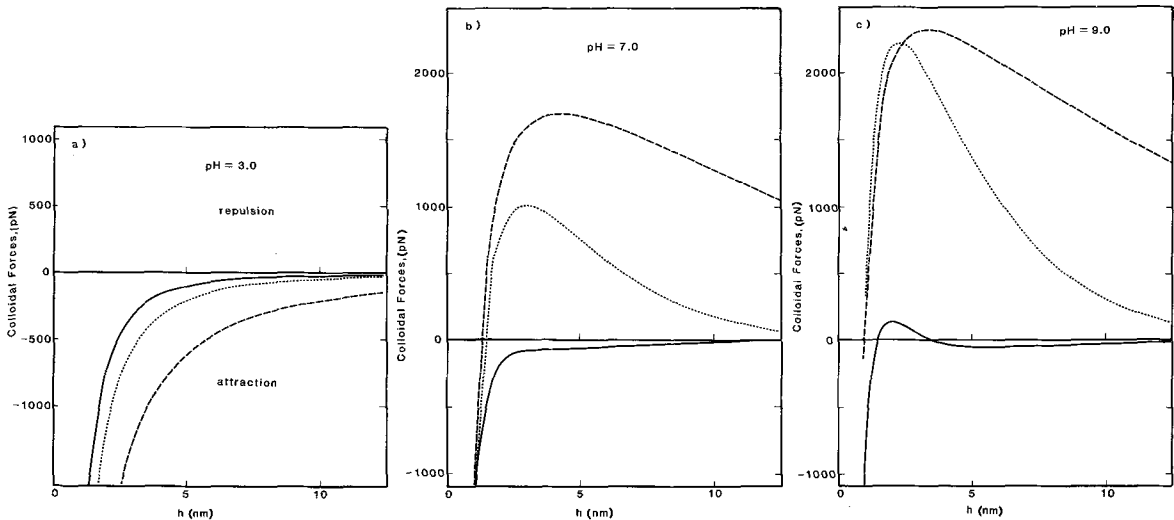


Figure 2. Interaction colloidal forces between a fine-grained particle and a sand grain surface. Computations made for (1) 10^{-1} M, (2) 10^{-2} M, and (3) 10^{-3} M NaCl.

tials had different signs according to Tables 1 and 2. As the pH increased, the colloidal forces changed drastically as a result of the magnitude of the zeta potentials. This change was especially evident at pH 7 (neutral conditions), at which van der Waals forces predominated only at high electrolyte concentrations; at low electrolyte concentrations, a repulsive force barrier was maintained. At pH 9.0, the active forces were mainly repulsive, except at 10^{-1} M, at which a slight repulsion barrier existed, followed by a secondary minimum as the separation distance increased.

Mobilization of fines

By comparing the magnitude of the colloidal forces (Figure 2) and the hydrodynamic forces, the mobilization of fines can be determined mainly by the chemistry of the formation. This effect is especially evident in Figure 3, in which the colloidal forces are plotted against $\log C$ for a given closest-separation-distance h_0 . As shown in this figure, the attractive forces changed to repulsive colloidal forces as the electrolyte concentration decreased. In fact, a critical electrolyte concentration (C_{cr}) existed, which depended solely on pH of the aqueous fluid. These C_{cr} values were evaluated from data such as those given in Figure 2 and are plotted against pH in Figure 4. Figure 4 was obtained from Figure 3 assuming a minimum separation distance of 20 Å. Figure 4 actually represents a mobilization diagram, which suggests that at a given pH, for formation salinity above the line, fines will not be mobilized, whereas for reservoir salinity below this line, fines will be mobilized. The same is true for the mobilization of kaolinite fines (Cerde, 1987), whose zeta potentials under neutral conditions are close to those of fines. Ac-

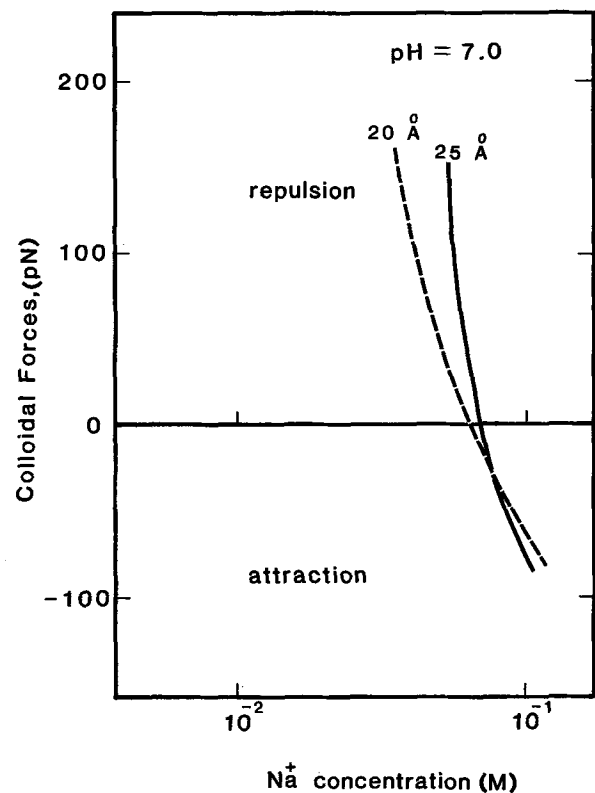


Figure 3. Interaction colloidal forces between a fine-grained particle and a sand grain surface as a function of NaCl concentration.

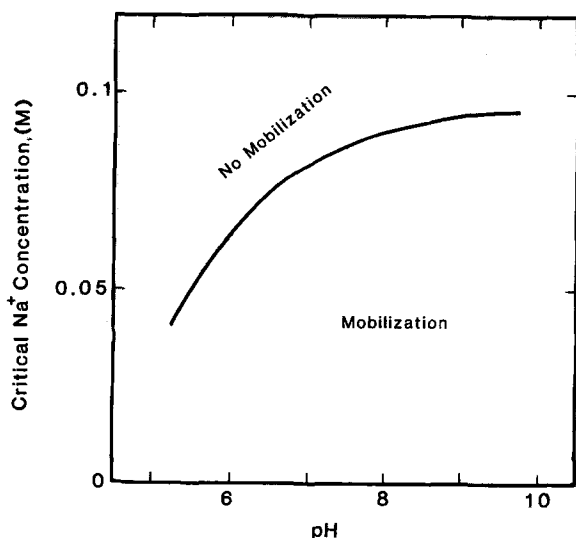


Figure 4. Mobilization diagram of fine-grained particle.

According to Cerda's analysis, under neutral conditions fines should mobilize at a NaCl concentration of 0.08 M, close to the value for the mobilization of kaolinite fines (Cerda, 1987) and in close agreement with experimental results reported by Khilar and Fogler (1981). At $\text{pH} > 8$ the critical Na^+ concentration showed little or no increase with increasing pH, because the zeta potentials of fines samples did not vary significantly for $\text{pH} > 8$.

Particles may become detached if the repulsive double layer force is increased (by decreasing the electrolyte concentration). Fines were initially attached to the sand grain surfaces due to van der Waals forces, which dominated at high electrolyte concentrations. The attractive forces were countered by the repulsive double layer forces. The reversal of the colloidal forces (from attractive to repulsive) explains the reduction in hydraulic conductivity reported by Goldenberg (1985) and Goldenberg *et al.* (1984), if the salinity of the injected water was abruptly changed from that of seawater (0.5 M NaCl) to that of aquifer freshwater of

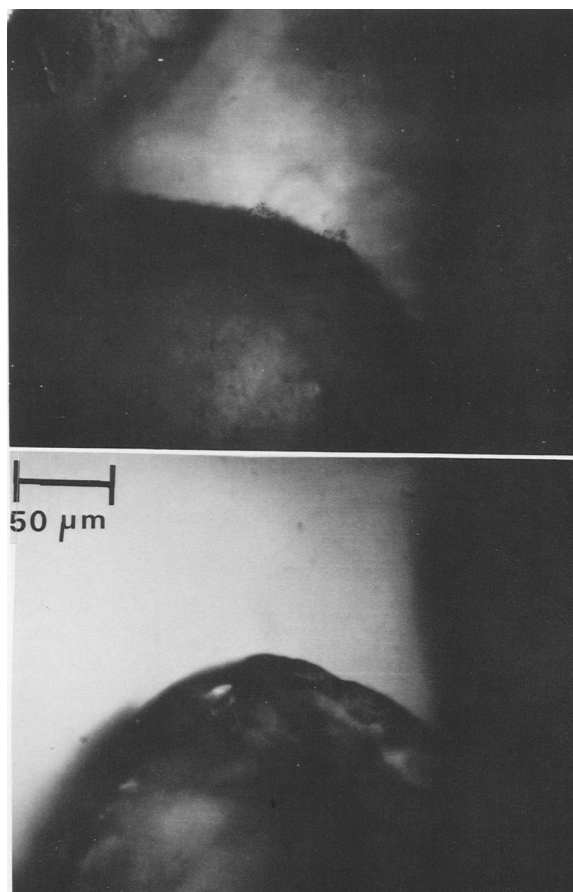


Figure 5. Photomicrograph showing the deposition of fine-grained particles on sand grain surfaces at pH 7.0. In upper micrograph, deposition occurred at high electrolyte concentration (10^{-1} M NaCl), whereas at low electrolyte concentration (10^{-3} M NaCl) deposition was not observed (lower micrograph).

very low salinity (10^{-3} M NaCl). As pointed out above, the reversal of the net attractive forces probably led to an increase in the separation distance between the fines and sand grain surface, and eventually the fines became detached by the flowing fluid. The effect of pH is quite evident in Figure 4. The C_{cr} were lower at low pH because the zeta potential values were lower than at high pH.

The above analysis predicts that for petroleum reservoir formations and aquifers containing high salinity fluids, fine-grained particles will not mobilize, regardless of their size. This is because under such conditions van der Waals attractive forces predominate. The effect of salinity decrease is evident in Figure 3, in which the mobilization of fines appears to have depended only on the chemistry of fluids present in the formations. As soon as water or steam is injected, mobilization

Table 3. Summary of the visualization experiment.¹

NaCl concentration (M)	pH 3	pH 7	pH 9
10^{-1}	Deposition	Deposition	Little deposition
10^{-2}	Deposition	Little deposition	No deposition, entrapment
10^{-3}	Deposition	Minimal deposition	No deposition, entrapment

¹ Deposition of quartz fines on Fisher silica sand (grade S-150).

should occur at a specific electrolyte concentration, as shown in Figure 4.

Visualization experiments

Experiments to observe the degree and manner of attraction of fines to the sand grain surface at different electrolyte concentration and pH are summarized in Table 3. At pH 3.0, the fines suspensions used were unstable and aggregates formed. Both single particles and aggregates were deposited on the sand grain surfaces (multilayer deposition occurred as well), as predicted by the model shown in Figure 2, in which attractive colloidal forces predominate independent of the electrolyte concentration. During these experiments, downward injection resulted in the deposition of fines on the top and lateral surfaces of the sand grains, but no deposition on the bottom surfaces of the sand grains, indicating that gravity was important. Under neutral conditions (pH 7.0), the deposition of fines occurred only at relatively high electrolyte concentrations. At a low electrolyte concentration ($C = 10^{-3}$ M NaCl) practically no deposition was observed, in good agreement with the theory, as shown in Figure 2b. At relatively high electrolyte concentrations the attractive colloidal forces between sand grain surfaces and fines should predominate, whereas at lower concentrations, fines must overcome a "force barrier" in order to be deposited.

Figure 5 shows the two extremes: the deposition of fines at an electrolyte concentration of 10^{-1} M NaCl, and no deposition at a concentration of 10^{-3} M NaCl. As mentioned above, fines were deposited only on the top surfaces of the sand grains if the fines suspensions were injected downward. At pH = 9.0, the fines suspensions were stable, and practically no aggregates were observed. At a relatively high electrolyte concentration, the deposition of fines was minimal, and only about a monolayer of fines was noted on the sand grain surfaces. The deposition of fines was not uniform, however, and regions in which no fines were deposited were also present. This heterogeneity was probably the result of two mechanisms of deposition:

- (1) Fines were deposited around the stagnation points, e.g., where the hydrodynamic forces acting on a fines particle were maximized.
- (2) Due to surface irregularity of the sand grain, the colloidal interaction forces were not the same from one point on the surface to another for the same separation distance.

These observations corroborated the types of interactions predicted by DLVO theory.

SUMMARY AND CONCLUSIONS

An analysis of the forces that act on fine-grained particles attached to sand grain surface in a sandpack was carried out to determine the critical conditions for

fines mobilization. The analysis has shown that if the displacing fluid is an aqueous solution, the mobilization of fine-grained particles is dictated mainly by the chemistry of the injected fluids. The mobilization occurs at certain electrolyte concentration, which depends on the pH of the fluids. The effect of fines size on the critical electrolyte concentration appears to have been practically negligible. If the displacing fluid was a viscous oil, the hydrodynamic forces predominated and the fines mobilization was determined by the velocity and the viscosity of the displacing fluids.

The present analysis may have importance not only in problems related to fines migration in petroleum reservoirs but also in sand control operations near the well bore and also in aquifers when freshwater replaces the seawater initially present in the formations.

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