Delft University of Technology

Physico-Chemical Factors of Clay Particle Migration and Formation Damage

Tchistiakov, A. A.; Kottsova, A. K.; Shvalyuk, E. V.; Zitha, P. L.J.

Important note
To cite this publication, please use the final published version (if applicable).
Please check the document version above.

Physico-Chemical Factors of Clay Particle Migration and Formation Damage

A. A. Tchistiakov[a], *, A. K. Kottsova[a], **, E. V. Shvalyuk[a], ***, and P. L. J. Zitha[b], ****

[a] Center for Hydrocarbon Recovery, Skolkovo Institute of Science and Technology, Moscow, 121205 Russia
[b] Delft University of Technology, Delft, the Netherlands
*e-mail: A.Tchistiakov@skoltech.ru
**e-mail: A.Kottsova@tudelft.nl
***e-mail: Elizaveta.Shvalyuk@skoltech.ru
****e-mail: p.l.j.zith@tudelft.nl

Abstract—This article describes the effects of different physico-chemical factors on formation damage caused by migration of in situ clay particles as a result of water injection into a clastic reservoir.

INTRODUCTION

Reduced well injectivity is a common problem with water intake in oil and geothermal fields. One of the most common causes of reduced permeability of rocks in the near-wellbore is clogging of the pore space as a result of in-situ clay particle migration, initiated either by a high injection rate or changes in physical and chemical conditions when a solution with a chemical composition different from the natural composition of formation water penetrates the formation. Despite the relatively active study of permeability loss problems in the last 2 decades (Tchistiakov, 2000; Bennion, 2002; Bedrikovetsky, 2014; Kalantarias, 2014; Kozhevnikov, 2016; Mikhailov et al., 2016; Civan, 2016), theoretical and experimental studies aimed at design of reservoir pressure maintenance systems and injection of low mineralized water to increase oil recovery in depleted fields, as well as development of geothermal reservoirs, remain highly relevant.

The aim of our work is a comprehensive theoretical analysis of the influence of physical and chemical factors on the migration of clay particles and experimental modeling of their migration process in a terrigenous reservoir.

THE THEORY OF CLAY PARTICLE MIGRATION AND PORE SPACE CLOGGING

The migration mechanism of clay particles and pore space clogging. A simple way to explain the mechanism of clay particle migration in rocks is to present a model of the mineral matrix of a relatively pure sandstone as a system of interconnected sand grains with dispersed clay particles and microaggregates adsorbed on the surface. The theoretical model of the rock pore space can be represented, accordingly, as a system of pores interconnected by narrow channels (Priisholm et al., 1987).

The physical and chemical properties of the pore fluid changed as a result of injecting an external solution may trigger disintegration of microaggregates and detachment of clay particles and microaggregates from sand grains. As a result, they are carried by the fluid and redeposited in narrower pore channels, which leads to a significant reduction in rock permeability in the near-wellbore (Vaidya and Fogler, 1990). The processes of microaggregate disintegration and clay particle desorption from the surface of sand grains are determined by colloidal interactions. Accordingly, it is logical to distinguish this mechanism of permeability loss as a separate colloidal type of formation damage (Tchistiakov, 2000).

Under the conditions of intense fluid injection, detachment and redeposition of clay particles and microaggregates can be initiated even without changing the physical and chemical properties of the fluid. This type of permeability loss according to the physics of the prevailing process is hydrodynamic (Vaidya and Fogler, 1990; Bennion, 2002). The degree of rock colmatation due to hydrodynamic particle migration depends on the fluid filtration rate, as well as the morphology and surface charge of clay minerals and sand grains. The surface charge is determined by the mineral composition of the sand and clay particles as well
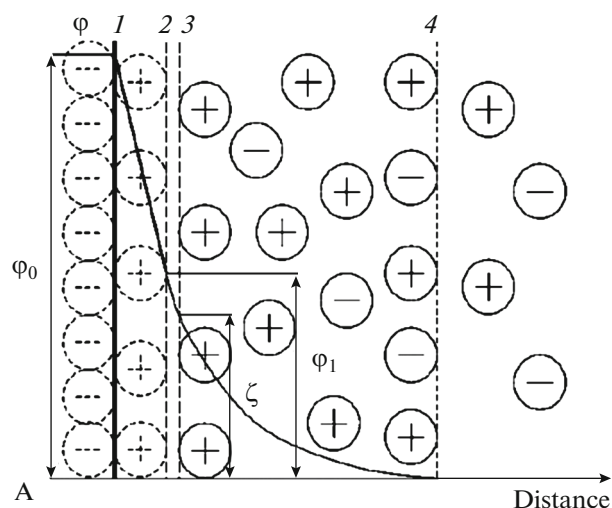
Fig. 1. The structure of the double electric layer on the boundary of clay particle with aqueous solution; φ_0, surface potential; φ_1, Stern potential; ζ-potential: (*1*) surface of clay particle; (*2*) boundary of adsorption layer; (*3*) sliding plane; (*4*) boundary of diffusion layer.

as by the physical and chemical properties of the pore solution.

The major physical and chemical factors affecting colloidal colmatation include the mineral composition of the rock and the concentration and composition of salts in the injected solution, as well as its pH and temperature (Fogler, 1990; Vaidya, 1990; Rosenbrand et al., 2015). Accordingly, the influence of each of these factors is considered in detail in the theoretical sections of the present article. To simplify the description of the migration mechanism, the general term "clay particles" is hereafter used for both microaggregates and individual particles, since such a division is often of purely theoretical significance anyway.

The formation of an electrical double layer around clay particles in aqueous solution. Isomorphic substitutions of Si^{4+} cations for cations of lower valence (Al^{3+}, Mg^{2+}, etc.) in the crystal structure of clay minerals lead to the formation of excess negative charge on their surfaces, which is compensated by positively charged exchange cations (Osipov and Sokolov, 1978, 2013; Kumar et al, 2017).

The exchange cations that occur in aqueous solutions are hydrated, which leads to the formation of the double electric layer (DEL) around the clay particles. The DEL consists of a negatively charged surface of the clay particle and a layer of cations with opposite charge in the surrounding solution. The layer of DEL cations, in turn, consists of an adsorption layer immediately adjacent to the surface of the clay particle, and a diffusion layer formed at some distance from the mineral surface as a result of cations dissociation in the aqueous medium. The cations of the adsorption and diffusion layers are attracted to the surface of clay par-

ticles by electrostatic forces and balance their surface negative charge (Fig. 1).

The ξ-potential is the potential difference between the dispersion medium and the stationary liquid layer surrounding the particle. The interaction between charged particles is determined primarily by the overlap of their diffuse layers; therefore, the ξ-potential is an important parameter to determine the type of interaction between particles, namely, their tendency to coagulate or disperse.

The charge of shear rupture in aqueous medium is formed as a result of its hydration and formation of SiOH-groups:

$$2(-SiO) + H_2O \rightarrow 2(-SiOH). \tag{1}$$
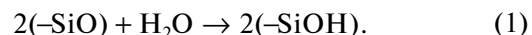
The positive or negative (depending on the pH of the medium) charge formed on the ruptures determines the stability of clay particles (Tchistiakov et al., 2001). This effect is described in more detail in the respective section.

The balance of forces acting on the clay particle adsorbed on the surface of the sand grain. The stability of the clay particle, i.e., its adsorption or desorption from the surface of the sand grain, is controlled by the balance of the following forces (Tchistiakov et al., 2000):

$$F_{tot}(h) = F_{LVA}(h) + F_{DLR}(h) + F_{BR}(h) + F_{HR}(h) + F_{IHL}, \tag{2}$$

where F_{tot} is the resultant force of interaction of the clay particle with the surface of the mineral matrix (it has a positive sign if repulsive forces predominate and a negative sign if attractive forces predominate); F_{LVA} is the London-Van der Waals force; F_{DLR} is the electrostatic repulsion force caused by interaction of the mineral matrix surface with DEL of the clay particle carrying the charge of the same sign; F_{BR} is the Born repulsion force (it has a significant effect at distances less than 5 Å); F_{HR} is the hydrodynamic force (this depends on the fluid velocity in the rock); F_{IHL} is the force of interaction of bound water layers occurring between the clay particle and the surface of the mineral matrix (this has a wedging effect); and h is the distance between the particle and the surface of the mineral matrix. Bedrikovetsky and Caruso (2014) proposed consideration of the gravitational force in the equilibrium equation. We note that its contribution to the balance of the acting forces is determined by the mutual orientation of the clay particle and the surface of the sand grain. According to (You et al., 2015), the gravitational component is 2–4 orders of magnitude smaller than the hydrodynamic and electrostatic repulsive forces.

The influence of pore solution mineralization. The electrokinetic potential is one of the key parameters determining the stability of clay particles and the total balance of their interaction forces with the surface of the mineral matrix (Khilar and Fogler, 1987; Rosen-

brand et al., 2015). Theoretically, if other parameters (e.g., temperature and pH) remain constant, an increase in the concentration of ions in the solution causes the transition of some cations from the diffusion layer to the adsorption layer due to the concentration gradient. This in turn leads to a decrease in the ξ-potential of the particles (Osipov et al., 1989).

Decreasing salinity and, consequently, an increase in the ξ-potential leads to increasing electrostatic repulsion between clay particles and the sand grains to which they were adsorbed during sedimentation. Consequently, pumping desalinated water into sandy reservoirs can cause clay particles to detach from mineral walls. The subsequent migration of clay particles together with the moving flow may lead to their redeposition in narrower pore channels, clogging the latter and, consequently, reducing the permeability of the rock as a whole (Davis et al., 1978; Vaidya and Fogler, 1990; Tchistiakov et al., 2001).

The influence of the pH of the pore solution. The charge of clay particles may vary depending on the pH of the aqueous medium (Davis et al., 1978; Osipov and Sokolov, 1978; Tchistiakov et al., 2001). In accordance with the existing ideas, partial dissociation of the hydroxyl and its transfer into the solution takes place in an alkaline environment, with an excessive negative charge being formed on the edges:

$$(-SiOH) \rightarrow SiO^- + H^+. \tag{3}$$
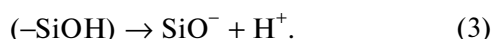
The $(-AlOH)^-$ group where Si^{4+} ion is replaced with Al^{3+} ion is also capable of dissociation depending on the pH. In an alkaline environment, the group dissociates to form a proton:

$$Al(OH)_3 \Leftrightarrow Al(OH)_2O^- + H^+. \tag{4}$$
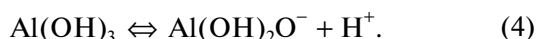
In an acidic environment, dissociation results in the release of the OH^- ion

$$Al(OH)_3 \Leftrightarrow Al(OH)_2^+ + OH^-. \tag{5}$$
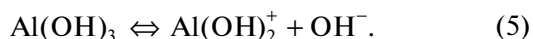
Thus, the ratio of OH^- and H^+ ions in solution affects the type and degree of dissociation and, therefore, determines the charge of the lateral chip. An increase in pH (i.e., an increase in the concentration of OH^- ions) leads to an increase in the negative charge on the side chips of clay particles. If the rock-forming mineral is quartz, whose surface charge is negative under natural conditions, an increase in pH above the isoelectric point of clay particles will cause their detachment from the surface of sand grains and, consequently, a decrease in rock permeability (Kottsova, et al., 2021). A decrease in pH below the isoelectric point induces a positive charge on lateral clay chips due to the protonation effect, which, in turn, contributes to the stabilization of clay particles on quartz grains, where the surface charge turns to positive at pH < 3.5, which is not typical of known reservoirs (Kottsova, et al., 2021).

For a long time it was believed that only lateral chips of clay minerals were subject to recharge. It has been relatively recently established that the basal surfaces of clay particles are also affected by pH, and the effect of pH increase may differ for silicate (tetrahedral) and aluminum (octahedral) lattices. In particular, it was found that silicate kaolinite surfaces acquire a negative charge at pH > 4, while aluminum surfaces are negatively charged only at pH > 9 (Kumar et al., 2017). Perhaps the difference in the isoelectric points of the tetrahedral and octahedral layers can explain the significant differences in the published isoelectric point values obtained for the same mineral.

The influence of temperature. An increase in temperature leads to an increase in the ξ-potential, which in turn contributes to the dispersion of particles (Rodríguez and Araujo, 2006; Rosenbrand et al., 2015). At the same time, the hydrodynamic effect on the adsorbed particle is weakened due to a decrease in fluid viscosity. Moreover, the wedging effect of the bound water layer also decreases (Tchistiakov, 2000a, 2000b). The published data suggest that the total effect of temperature still contributes to desorption of clay particles, their migration and subsequent reduction in the rock permeability (Nozdratenko, 2015; You et al., 2015). However, when interpreting the experimental data, it should be born in mind that the temperature-induced permeability decrease may also be due to thermal expansion of the rock matrix and closure of pore channels (Cheng and Milsch, 2020).

MATERIALS AND METHODS

In order to study the effect of decreasing salinity on the permeability of a terrigenous reservoir, a series of filtration experiments was performed on sandstone samples from the Bentheimer Formation. X-ray analysis revealed that the sandy fraction of the rock is represented almost entirely by quartz while the clay fraction was mostly kaolinite. The proportion of quartz and kaolinite in the total composition of the rock was 98 and 2%, respectively. The length of the sample was 17 cm, diameter 3.8 cm. To simulate the effect of decreasing salinity on migration of clay particles, a NaCl solution with salinity of 5 g/L the sample was soaked in at first, was replaced with demineralized water during the experiment.

The pressure was measured during the experiment along the sample at three equal intervals to monitor permeability variations during fluid filtration. A 0.5 μm filter was installed in front of the core sample holder in order to filter out impurities from the injected solution. The fluid exiting the sample was collected into cuvettes for subsequent analysis of suspended solids. The volume of each sample was 0.2 of the total volume of the sample pores. The turbidity of the filtrate was measured by nephelometry, and the size distribution of suspended particles was determined by dynamic light scattering.

Table 1. The parameters of the experiment

Stage of experiment	1	2	3	4	5	6
Demineralized water flow rate, mL/min	1.18	2.36	4.72	7.08	9.95	11.81

The experiment consisted of several stages: saturation of the core with saline solution, permeability measurement, injection of demineralized water with different flow rates, and one more permeability measurement. The injection parameters are shown in Table 1. The volume of injected fluid was equal to ten pore volumes at each stage. In this article, we present the results of one of the most representative tests; other experiments showed similar patterns.

RESULTS AND DISCUSSION

The measured permeability of the sample initially saturated with sodium chloride solution are given in Table 2. The close values of permeability in all sections of the sample indicate its homogeneity, which allows a comparative analysis of differential pressure values measured during the subsequent filtration of demineralized water.

Upon replacing the sodium chloride solution with demineralized water, a pressure drop was recorded consistently in all sections of the sample at the first flow rate step, equal to 1.18 mL/min (Fig. 2). Suspended particles were detected in the filtrate immediately after replacement of the first pore volume by demineralized water (Fig. 3). We note that their concentration gradually increased, reaching its maximum after filtering five pore volumes of water, after which it decreased and reached its minimum when the last, tenth, pore volume was pumped through the sample.

Increasing the demineralized water flow rate to 2.36 mL/min at the second stage of the experiment led to an increase in the pressure drop in all intervals of the sample, starting approximately from the fourth pore volume that was pumped through. There are two peaks on the turbidity plot (Fig. 3). The first peak corresponds to the passage of the first pore volume at the increased flow rate. The second surge in the concentration of suspended particles in the filtrate begins after passing the fourth pore volume, which corresponds to the previously mentioned increase in the pressure drop (Fig. 4). On the basis of these data, one can assume that the flow rate of 2.36 mL/min, corresponding to a linear flow rate of 0.2 cm/min, exceeds the critical value for kaolinite particles in the studied sandstone. This resulted in their hydrodynamic detachment from the surface of the mineral matrix and a decrease in rock permeability. It is important to note that after each successive stage of fluid flow the observed pressure drop in the outlet section of the core was becoming increasingly significant compared to those in the inlet and middle sections of the sample, which can be explained by the gradual accumulation of particles migrating from the first two sections. The calculated values of permeability decrease compared with the initial one (Table 2) for three sections of the sample also indicate the regular decrease in the permeability in the direction of liquid flow.

Analysis of filtrate turbidity showed that each increase in fluid flow rate above the critical one (2.36 mL/min) caused detachment of additional clay
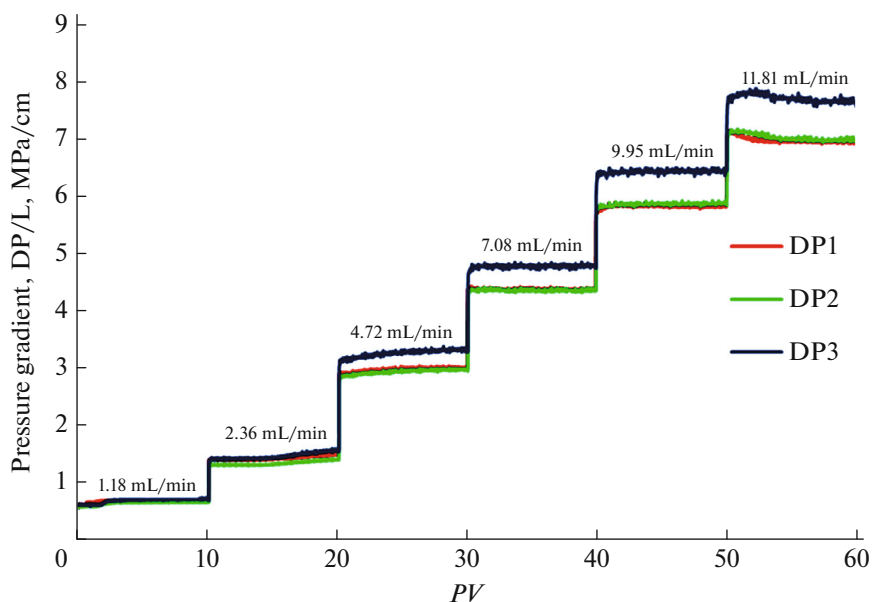
Fig. 2. The differential pressure gradient as a function of pumped liquid volume at different phases of the experiment.
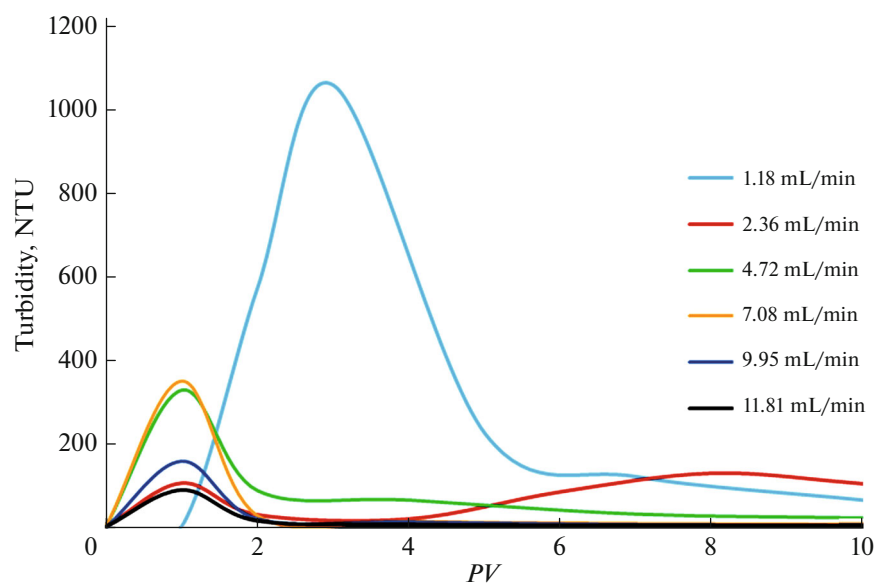
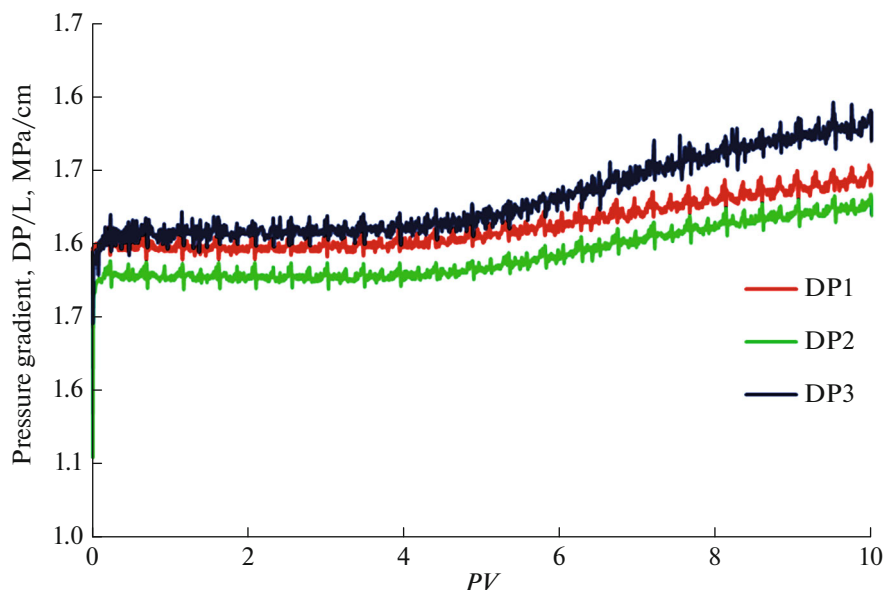**Fig. 3.** The turbidity of the solution flowing out of the core at different stages of the experiment.

Fig. 4. The pressure drop gradient at the stage of demineralized water injection at a flow rate of 2.36 mL/min.

particles and an increase of filtrate turbidity, which decreased in the process of passing several pore volumes of water. At the same time, the peak concentration of turbidity of filtrate was observed at the moment of release of the first pore volume of liquid. Starting from the fourth flow rate of 7.08 mL/min, kaolinite particles were recorded only in the first 2.3 pore volumes of filtered water and the total amount

Table 2. The core permeability before and after filtration experiments

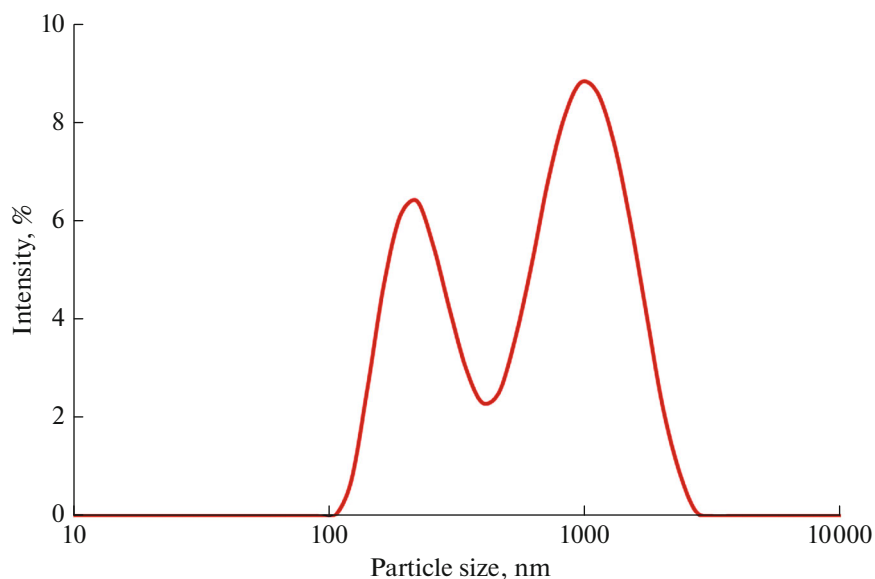| Section	Initial core permeability, $\mu m^2 \times 10^3$	Final core permeability, $\mu m^2 \times 10^3$	Permeability decrease, %
Total	1900	1600	15
1	1780	1660	7
2	1930	1620	16
3	860	1510	19

Fig. 5. The kaolinite particle size distribution.

of particles carried out of the sample decreased after each flow rate step.

Analysis of the size of the dispersed kaolinite particles in the filtrate showed the presence of two peaks corresponding to particle diameters of 0.2 and 1 μm. The maximum diameter of suspended particles was about 5 μm. Accordingly, all particles of larger diameter retained in the rock. According to the mercury porosimetry data, the diameters of the pore channels in the rock range from 10 to 30 μm (Fig. 5).

CONCLUSIONS

(1) Reduction of mineralization from 5 to 0 g/L NaCl initiates migration of internal clay particles and, as a consequence, leads to reduction of rock permeability. The degree of permeability reduction increases in the direction of fluid movement from the inlet to the outlet section of the sample due to clogging of pore channels by particles moving with the flow.

(2) It was established that filtrate turbidity gradually increases after the replacement of sodium chloride solution with demineralized water, reaches its maximum after filtering first five pore volumes of water, and decreases afterwards. In principle, a gradual increase in the concentration of suspended solids in the filtrate can be explained by a combination of several factors, namely nonsimultaneous detachment of clay particles from sand grains, the different time when they accelerated to the speed of fluid flow, and differences in their size, shape, and therefore, hydraulic size, as well as different water flow rates at the microlevel and partial retention in pore constrictions.

(3) The critical flow rate of demineralized water for kaolinite particles in the Bentheimer Formation sand-stone of 2.36 mL/min corresponds to linear flow rate of 0.2 cm/min. This value was established based on measurements of pressure drop along the core sample and analysis of the turbidity of the output filtrate.

(4) Each increase in flow rate above the critical value leads to detachment of an additional portion of kaolinite particles due to the influence of hydrodynamic force. The amplitude of the turbidity peaks of the filtrate decreases with each subsequent surge of demineralized water.

(5) The comparative analysis of the turbidity plots obtained for the studied rock suggests that the electrostatic repulsion that increased as a result of a salinity drop caused migration of more clay particles compared to an increase in the hydrodynamic flow rate.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

REFERENCES

Bedrikovetsky, P. and Caruso, N., Analytical model for fines migration during water injection, *Transp. Porous Media*, 2014, vol. 101, no. 2, pp. 161−189.

Bennion, D.B., An overview of formation damage mechanisms causing a reduction in the productivity and injectivity of oil and gas producing formations, *J. Can. Petrol. Technol.*, 2002, vol. 41, no. 11, pp. 29−36.

Cheng, C. and Milsch, H., Permeability variations in illite-bearing sandstone: Effects of temperature and NaCl fluid salinity, *J. Geophys. Res. Solid Earth*, 2020, vol. 125, no. 9.

Civan, F., *Reservoir Formation Damage*, Thirded, 2016.

Davis, J.A., James, R.O., and Leckie, J.O., Surface ionization and complexation at the oxide/water interface: I. Computation of electrical double layer properties in simple electrolytes, *J. Colloid Interface Sci.*, 1978, vol. 63, no. 3, pp. 480−499.

Kalantariasl, A. and Bedrikovetsky, P., Stabilization of external filter cake by colloidal forces in a "well-reservoir" system, *Ind. Eng. Chem. Res.*, 2014, vol. 53, no. 2, pp. 930−944.

Khilar, K.C. and Fogler, H.S., Colloidally induced fines migration in porous media, *Rev. Chem. Eng.*, 1987, no. 4, pp. 41−108.

Kottsova, A.K., Yegane, M., Tchistiakov, A.A., and Zitha, P.L.J., *Effect of Electrostatic Interaction on the Retention and Remobilization of Colloidal Particles in Porous Media*. Moscow, 2021.

Kumar, N., et al., Probing the surface charge on the basal planes of kaolinite particles with high-resolution atomic force microscopy, *Langmuir*, 2017, vol. 33, no. 50, pp. 14226−14237.

Kozhevnikov, A.V., Influence of mineralization of reservoir waters on permeability and oil recovery of reservoirs, *Nauchn. Forum. Sibir', Estestv. Nauki*, 2016, no. 3, pp. 64−66.

Mietta, F., Chassagne, C., and Winterwerp, J.C., Shear-induced flocculation of a suspension of kaolinite as function of pH and salt concentration, *J. Colloid Interface Sci.*, 2009, vol. 336, no. 1, pp. 134−141.

Mikhailov, D.N., Ryzhikov, N.I., and Shako, V.V., Experimental investigation of transport and accumulation of solid particle and clay suspensions in rock samples, *Fluid Dynamics*, 2015, vol. 50, pp. 691−704.

Mitchel, J.K., *Fundamentals of Soil Behaviour*, John Wiley, Inc. New York, 1976.

Nozdratenko, S.A., Akul'shin, A.A., and Pereverzeva, V.S., Types of wall packing, *Pokolenie Budushchego: Vzglyad Molodykh Uchenykh*, 2015, pp. 163−165.

Osipov, V. and Sokolov, V., Structure formation in clay sediments, *Bull. Assoc. Eng. Geol.*, 1978, vol. 18, pp. 83−90.

Osipov, V.I. and Sokolov, V.N., *Clays and Their Properties. Composition, Structure and Formation of Properties*, Moscow: GEOS, 2013.

Osipov, V.I., Sokolov, V.N., and Rumyantseva, N.A., *Mikrostruktura glinistykh porod* (Clay Rock Microstructure), Moscow: Nedra, 1989.

Priisholm, S., Nielsen, B.L., and Haslund, O., Fines migration, blocking, and clay swelling of potential geothermal sandstone reservoirs, Denmark, *SPE Form Eval.*, 1987, vol. 2, no. 2, pp. 168−178.

Rodriguez, K. and Araujo, M., Temperature and pressure effects on zeta potential values of reservoir minerals, *J. Colloid Interface Sci.*, 2006, vol. 300, no. 2, pp. 788−794.

Rosenbrand, E., et al., Different effects of temperature and salinity on permeability reduction by fines migration in Berea sandstone, *Geothermics*, 2015, vol. 53, pp. 225−235.

Tchistiakov, A.A., Colloid chemistry of *in-situ* clay-induced formation damage, in *SPE Int. Symp. Form. Damage Control. Lafayette*, 2000a, pp. 371−379.

Tchistiakov, A.A., Physico-chemical aspects of clay migration and injectivity decrease of geothermal clastic reservoirs, in *Proc. World Geotherm. Congr., Kyushu, Tohoku, Japan, May 28−June 10*, 2000b, pp. 3087−3095.

Vaidya, R.N. and Fogler, H.S., Fines migration and formation damage, in *Influence of pH and Ion Exchange*, 1990, no. 11, pp. 125−132.

You, Z., Bedrikovetsky, P., Badalyan, A., and Hand, M., Particle mobilization in porous media: Temperature effects on competing electrostatic and drag forces: temperature effects on fines migration, *Geophys. Rev. Lett.*, 2015, vol. 42, no. 8, pp. 2852−2860.

Translated by M. Hannibal