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Physico-chemical factors of clay particle stability and transport in sandstone porous media

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ABSTRACT

Formation damage caused by clay particle transport is a world-wide problem in geothermal energy exploitation as well as in oil industry. In this paper we give a theoretical overview over physico-chemical factors determining stability and transport of clay particles in sandstone reservoirs.

KEYWORDS

Geothermal energy, formation damage, clay particle transport, physical chemistry.

Introduction

Clay induced formation damage during water re-injection can be caused by external as well as by internal clays released from the rock pore walls. The rate of permeability decline depends on the intrinsic fluid velocity and physico-chemical factors controlling the electrical double layer around clay particles.

The quantitative theory of colloidal particle interactions was developed by Derjaguin B.V. (1937), Landau L.D. (1941); Verwey J.J.W. and Overbeek J.Th.G. (1948) (DLVO theory). Later on, based on the DLVO theory principles, the theory of clay particle interaction and clay rock properties was developed (H. van Olphen, 1963; J.K. Mitchel, 1976; V. I Osipov, 1979). It is well known now, that attraction Van der Waals-London forces with repulsion electrostatic forces and structural forces of adsorbed water layers control the stability of clay particles in water suspensions and sandstone reservoirs.

In this paper we highlight the mechanisms of formation of clay surface charge and the clay particle electrical double layer (EDL). Then we consider the influence of physico-chemical factors on coagulation - dispersion processes and the rate of pore throat bridging by internal clays. In particular we describe the effect of the ionic strength, chemical composition, pH, and the temperature of the permeating fluid, as well as clay mineralogy and composition of the exchangeable cations. Finally we describe the effect on clay stability of structural forces of adsorbed water layers which are often ignored in reservoir engineering literature. We believe that better understanding of the main theoretical principles of the clay particle behaviour will help the geothermal and petroleum specialists to develop better techniques for geothermal and oil reservoir permeability enhancement.

Mechanisms of clay surface charge formation and clay particle electrical double layer structure

Clay basal surfaces

The charge of a clay basal surface mainly depends on the isomorphic changes occurring in the clay crystal. For example, the Si^{4+} cation can be substituted by Al^{3+}, Mg^{2+}, etc. As a result the crystal structure gets a negative charge that is compensated by exchangeable cations in dry conditions. In presence of water (e.g. in aqueous suspension) the exchangeable cations dissociate, thus the clay crystal surface gets negatively charged and the electrical double layer (EDL) is formed. Theoretically the EDL can be represented as a “condenser” where the negatively charged inner layer of the condenser is built up by the clay surface and positively charged outside layer is formed by the exchangeable cations.

Clay edges

When a clay crystal edge interacts with water, (-SiO) -groups are hydrated and form (-SiOH)-groups on the crystal edge surface: $2(\text{-SiO}) + H_2O \rightarrow 2(\text{-SiOH})$. In base conditions the formed poly-silicon acid partially dissociates: $(\text{-SiOH}) \rightarrow SiO^- + H^+$. The produced H^+ ions (or H_3O^+) transit to the aqueous phase, but under the electrostatic attraction of (-SiO^-)-groups they remain nearby the crystal surface.

The concentration of OH and H^+-ions in the solution effects the degree of the (-SiOH) - group dissociation and thus determines the potential of the silicon surface (i.e. clay edge charge). An increase of pH (i.e. concentration of the OH-ions) increases the silicon surface negative potential and thus stabilises clays in aqueous suspensions. The dispersion effect of diluted alkaline solutions can complicate the injection water purification from clays. In clayey sandstone it can lead to the detachment of internal clay particles from the sand grains and cause clay induced formation damage.

If Si^{4+} is isomorphically changed with Al^{3+}, the type of the (-AlOH)- group dissociation depends on the pH. In base conditions the (-AlOH) - group dissociates by the acid type ($Al(OH)_3 \leftrightarrow Al(OH)_2O^- + H^+$), and in acid condition - by the base type ($Al(OH)_3 \leftrightarrow Al(OH)_2^+ + OH^-$). In the second case the edges of the clay particles get positively charged.

In many minerals (e.g. in montmorillonites) the isomorphic exchange of Si^{4+} for Al^{3+} occurs only in octahedral layers. If there are no isomorphic exchanges for Al^{3+} in tetrahedral positions, the edges of tetrahedral layers in acid environment remain neutral and get negatively charged in base conditions. Nevertheless it is important to notice that in mica and hydromica isomorphic exchange of Si^{4+} for Al^{3+} can be present in tetrahedral layers as well, and thus the edges of these layers behave similar to octahedral ones.

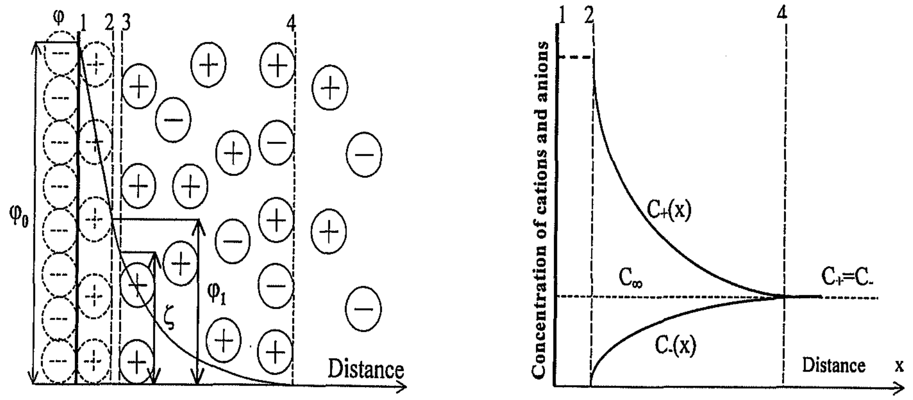
Besides described above there are also some other mechanisms of charge formation in clay minerals that are considered in details in clay mineralogy literature (Sergeev E.M. 1983).

Clay particle electrical double layer structure

In water environment the hydration reduces the strength of bonds between a clay surface and exchangeable cations (cations-compensators or counterions). While part of the cations remains attached to the clay surface and form the adsorbed cation (or so-called Stem) layer, another part of the cations transits at some distance from the clay surface and forms the diffuse ionic layer (figure 1, A). The distribution of the dissociated cations near a clay particle surface is determined by the balance between electrostatic attraction of the clay surface and thermal motion of the ions, tending to spread the ions away from the surface and equalise their concentration in the solution. Consequently the concentration of the dissociated cations decreases with the distance from the particle attaining the concentration of the similar cations in the solution (figure 1, B). The concentration of the anions on the contrary decreases in the direction towards the surface.

The clay surface charge is balanced by an equivalent number of oppositely charged cations in adsorption and diffusion layers. The part of the clay surface (or thermodynamic) potential, which is compensated by the adsorbed cation layer, is called consequently the adsorption potential. The remaining part of the potential, at the boundary between adsorbed and diffusion layers, is called Stem potential.

The interaction between charged particles is governed predominantly by the overlap of their diffusion layers, so the Stem potential is often considered as the most relevant to the interaction (Gregory. J, 1989). Nevertheless there is no direct methods for determining thermodynamic as well as Stern potentials. So far instead of the Stem potential the electrokinetic or ζ -potential is often used for calculations. The electrokinetic potential is that at the plane of shear, between a particle and a fluid, when there is relative motion between them (e.g. due to electrophoretic clay transport or electroosmotic fluid flow). Consequently the ζ -potential of clays can be found by means of electrophoresis or electroosmosis techniques. In spite on the fact that the shear plane does not coincide precisely with the adsorption layer boundary (the Stem plane), ζ -potential can be used as an adequate substitute of the Stem potential.



A B

Figure I: A principle scheme (Stern model) of the electrical double layer on the boundary clay basal surface - aqueous solution. A - cation and electrical potential distribution near a charged surface. B - cation (C_+) and anion (C_-) concentration as a function of a distance from the surface. φ_o - surface potential; φ_l - Stern potential; ζ - electrokinetic potential; 1 – clay surface; 2 – Stern plane; 3 - plane of shear, between a particle and fluid; 4 – boundary between a diffuse layer and free solution.

Effect of physico-chemical factors on ζ-potential, EDL properties, and stability of clay particles

The electrokinetic potential is one of the main parameters that determine the clay particle stability in suspensions and the strength of clay-mineral skeleton contacts in sandstone reservoirs. **An** increase of the <-potential adds to the electrostatic repulsion between clay particles and thus stabilises clay suspensions and complicates the injection water filtration. In a sandstone an increase of the ζ-potential of clay particles and the rock matrix grains can lead to release of the clays from the pore walls and cause pore throat plugging.

The value of ζ-potential and thickness of the diffuse layer are mainly controlled by the following factors: clay surface potential (i.e. mineralogy); **type** of the exchangeable cations, concentration and chemical composition of the pore solution; dielectric constant of the solution, pH and temperature. In the following paragraphs we consider the influence of these factors on the <-potential, EDL structure and stability of clay particles.

Clay mineralogy and composition of the exchangeable cations

The value of the surface potential of clay minerals generally increases with the number of hetero-valence isomorphic changes in their crystal structure. Nevertheless one should be aware that the excessive charge of the octahedral layers can be significantly screened by tetrahedral ones, e.g. in the minerals with 2 (tetrahedral layers) : 1 (octahedral layer) structure. However the montmorillonite minerals, because of a large specific surface, have better developed diffuse layers and exhibit rather larger swelling (and consequently swelling induced formation damage) relatively to kaolinite and hydromica.

Value of the clay <-potential significantly varies for different cation forms of the same mineral. Increase of valence of exchangeable cations strengthens bonds between the cations and the clay surface and consequently reduces the <-potentialand the diffuse layer thickness.

Polarisability of a cation increases and its hydration ability decreases with increasing of the cation radius. Larger polarisability and less hydration ability of a cation contribute to strengthening of its adsorption bonds with a clay surface. Therefore, if the exchangeable cations have the same valence, the increase of their radius should lead to decrease of ζ-potential and the diffusion layer thickness (except H^+).

These theoretical propositions were confirmed by numerous experimental investigations. According to Efiemov I. F. et al. (1983), for mono-ionic forms (Li, Na, Cs, H) of kaolinite the ζ -potential reduces in the following order:

Li (<=-71 mV) > Na (<=-48 mV) ≈ Cs (-49 mV) > H (-24 mV).

Many scientists claim that the surface charge of kaolinite is mainly concentrated on the edges of clay particles where the valence bonds are not filled. Because there are a few isomorphic changes in the kaolinite structure, the basal surfaces have relatively small charge and the exchangeable cations are mainly adsorbed on the crystal edges. This specific charge distribution distinguishes the kaolinite from most other clay minerals, which have higher values of ζ–potential on basal surfaces (Grim R.E. 1963, Osipov V.I. et al. 1985).

Zlotchevskaia R. I. and Korolev V. A. (1988) have found that <-potential also depends on the water saturation of the rocks. According to their electro-osmotic tests (which can measure only the averaged value of <-potential of clay basal surfaces and edges) <-potential changes from -10 to -50 mV for kaolinite, and from -15 up to -100mV for montmorillonite, depending on the moisture of the investigated samples.

F.D. Ovcharenco, B.E. Poliakov and O.L. Alekseev (1971) show that at equal concentrations of the pore solution the <-potential of different ionic forms of vermiculite decreases as follows: Li>Na>NH_4>Ca> Cu>Mn>Co, and for palygorskite: NH_4>Ca>Mn>Co>Cu.

J.P. Quirk and R.K. Schofield (1955), K.C. Khilar and H.S. Fogler (1987) found experimentally that a ***critical ionic strength,*** lower which clays start being released and transported in sandstone, for KCl solutions is rather low then for NaCl solutions of similar concentrations.

Most of the other experimental data also confirm that in general the clay stability in sandstone decreases with the decrease of the exchangeable cation charge (except H^+) and radius.

Effect of pH

As mentioned above the edges of clay minerals are negatively charged in alkaline solutions and positively charged in acid solutions. The value of the iso-electric point can vary significantly depending on the mineralogy and crystal structure of clays. According to V.I.

Osipov (1979), for montmorillonite the iso-electric point is equal to 6.5, for hydromica - 6 and for kaolinite - about 8. D. Williams and K. Williams (1977) found from electrophoretic measurements that the iso-electric point of kaolinite amounts to 7.4.

It is important that high concentration of some poly-valence cations in pore solution can also cause change of <-potential sign of natural minerals. For instance it is experimentally shown that surfaces of quartz grains get positively charged in highly concentrated solutions of $AlCl_3$ (Tchierniak A.S. et al., 1983).

Change of the charge sign at the clay edges to a positive one causes coagulation of clay suspensions and consequently simplifies the process of the injected water filtration. In acid environment clay suspensions form sediment with highly porous but strong microstructure where basal surface - edge type of contacts dominates. The strength of the contacts between the oppositely charged surfaces is much higher than the strength of the contacts formed only due to Van der Waals-London attraction in base conditions (Osipov V.I. 1979, Osipov V.I. et al. 1989, Tchistiakov A. A. 1994). In sandstone reservoirs with acid fluid in the porous medium the internal clays should be more stable relative to alkaline conditions, if the pore walls (e.g. quartz) remain being negatively charged and any other formation damage factor is not involved.

Effect of the salt concentration in the pore solution.

Here we consider the effect of salts, which do not change the surface potential of clay particles themselves (indifferent electrolytes), but effect the thickness of the EDL. Generally, if the other physico-chemical conditions (moisture, temperature, particle size, pH, surface potential) are constant, the increase of salt concentration causes reduction of the electrical double layer and a decrease of the <-potential (van Olphen H. 1963, Gregory J. 1989, Sergeev E.M. et al. 1983, Tchistiakov A. A. 1991). This is explained by the fact, that if the salt concentration in the pore solution increases, part of the cations move from the diffuse layer to the adsorption layer and consequently the <-potential decreases (for simplification we presume that the exchangeable cations and the cations in the pore solution are the same).

Nevertheless it is important to emphasise that some experiments show that replacement of distilled water in Na-forms of clays with diluted solutions of NaCl can cause increase of ζ-potential and enlargement of the diffusion ionic layer relative to distilled water. For different minerals the NaCl dispersing effect on clay soils and suspensions was observed at concentrations between 0.001 M and 0.01 M. (Zlotchevskaya R. I. et al., 1988, Tchistiakov A. A. 1994). For Na-form of the minerals a possible mechanism of this phenomenon can be the following. While only distilled water is present in a pore medium, part of H' diffuses to the exchange complex of clay minerals. The permeating of NaCl solution causes exchange of part of H^+ for Na', that leads to increase of the clay zeta potential (see above). Further increase of NaCl causes successive reduction of <-potential. A similar mechanism of the NaCl dispersion effect can be suggested for poly-valence forms of clay minerals.

Effect of cation exchange reactions on the clay stability

In practice we usually deal with aqueous solutions which contain several salts. The cations of the most common salts can be distributed in the following order according to their adsorption capacity (or energy of their adsorption at clays): $Fe^{3+} > Al^{3+} > H^{+} > Ba^{2+} > Ca^{2+} > Mg^{2+} > NH_4^{+} > K^{+} > Na^{+} > Li^{+}$ (Sergeev E.M. et al., 1983). Thus the mono-valence cations can easier desorb from a clay surface (except H^{+}) and go to the diffuse layer around the clay particle. It means, that if the concentration of the cations in the solution is equal, the equilibrium in the exchange reactions is always shifted to poly- valent cations.

The *critical coagulation concentration* of salts, above which clay suspensions coagulate, strongly depends on the valency of the cations, present in the solution. The value of the coagulation limit decreases with increase of the cation valency and consequently with the partial concentration of the poly-valent salts in the solution. That is why poly-valent salts are widely used to purify water from clays.

It is reasonable to suggest that salts, containing poly-valent cations, have a stabilizing effect on clay particles in sandstones due to strengthening of the clay-matrix bonds, and therefore can be considered as chemicals that counteract formation damage caused by in-situ clays. Khilar K. C. and Fogler H.S. (1987) experimentally proved this theoretical proposition. They have shown that the increase of the partial concentration of $CaCl_2$ in the mixture with NaCl decreases the value of the *critical ionic strength* of the solution permeating through the illite soil bed.

Effect of temperature

Based on the results of elecro-osmosis measurements in Na-forms of kaolinite and montmorillonite, Zlotchevskaia R.I. and Korolev V.A. (1988) show that the <-potential grows almost linearly when the temperature increases from 5° C to 70°C. The increase of the <-potential for kaolinite amounts to **40%** and for montmorillonite reaches **55%.** M. Fazilova (1976), M.P. Sidorova and D. A. Fridrikhsberg (1980) detected growing of the <-potential for quartz fibbers with increase of the temperature as well.

Based on these data it is possible to suggest that the increase of the temperature should increase stability of clay particle in suspensions. In sandstone the temperature increase therefore should stimulate clay particle dispersion from the mineral grains.

Nevertheless V. A. Korolev and **A. A.** Tchistiakov (1988) have experimentally found that the effect of temperature on stability of clay suspensions is much less evident than the effect of other physico-chemical factors. It depends on the mineralogy of clay particles, ionic strength of the solution as well as on the interval of the temperature. For example, they have shown that the increase of ionic strength of aqueous clay suspensions significantly reduces the effect of temperature on the coagulation processes.

On one hand the rise of the temperature increases the value the <-potential and contributes to dispersion of clay particles. On the other hand the temperature increase reduces splitting effect of bond water layers around clay particle (see below). Thus the total effect of the temperature on clay particles stability will depend on the dominance of one of these factors in particle-particle (or particle-mineral matrix) interactions.

Beside physico-chemical researches, effect of temperature on clay particles stability and formation damage was intensively investigated in reservoir engineering. Anuj Gupta and Faruk Civan (1994) give an extensive theoretical overview on temperature sensitivity of sandstones. Unfortunately there is still no robust agreement between experimental results and theory in clay sciences as well as in formation damage domain.

Structural forces of water films around clay particles

In aqueous environment clay minerals adsorb water molecules, and water films form on the particle surfaces (figure 2). Many authors distinguish two main structural water layers: adsorption (or "firmly bonded") water layer (a-film, after B. V. Derjaguin), and "osmosis" (or loosely bonded) water layer (β-film). These two types of water films are characterised by different physico–mechanical properties and strength of contacts with a clay surface.

Within the a-film we can theoretically distinguish two layers. The water layer, closest to a clay surface, is formed by molecules, which hydrate the clay crystal surface and adsorbed cations. The water molecules are firmly attached to the clay particle mainly due to hydrogen and ion-dipole bonds. The outer layers of the a-film are formed by water molecules, highly oriented toward the clay surface due to dipole-dipole interactions with the water molecules attached immediately to the clay.

The thickness of the a-film depends on crystallographic parameters of the clay particle and physico-chemical properties of the pore solution. B. V. Derjaguin (Derjaguin B.V., Churaev N.V., 1984) experimentally shows that for a pure silicon surface it can vary from 1.5 - 8 up to 30 nanometers. Because of the forced orientation of the water molecules within the a-film, the adsorbed water has higher values of viscosity and shear yield limit relative to "free" water. N.F. Bondarenko (1973) experimentally determined the a-film shear yield limit of 9.5-13 Pa. The shear yield limit of free water is rather less, and equal to 10^{-3} Pa. The dielectric constant of adsorbed water films in montmorillonite, which have a thickness of 5-8 nanometers, amounts to only 24 (while for water it is 81). Many other parameters of the adsorbed water, e.g. temperature of freezing and thermal conductivity, also significantly differ from properties of free water.

The β-film is a transition layer between highly structured water of the adsorption layer and free water. Within the β-film part of the water molecules is associated with the diffused exchangeable cations that cause distortions in the water structure. The exterior boundary of the osmosis water theoretically coincides with the outward boundary of the diffuse ionic layer. The physical properties of the osmosis and free water are comparable.

The structural forces associated with the water films have a splitting (repulsion) effect on the clay particles and thus the balance between coagulation-dispersion processes depends on the thickness of the films (B.V. Derjaguin, A.S. Titievskaia, 1953). Based on the experimental results of B.V. Derjaguin and N.V. Churaiev (1984), V.I.Osipov et al. (1989) show that the water film thickness at clay edges can be ten times as less as at the basal surfaces. This is a reason why clay particles in suspensions are less stable against coagulation on their edges (**A. A.** Tchistiakov, 1994). In sandstone clay microaggregates frequently interact with the rock matrix by their edges as well.

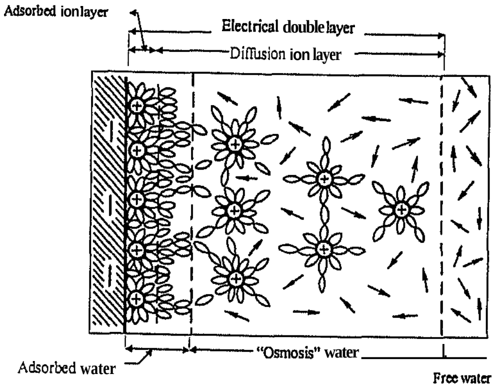


Figure 2: Structure of water films near a clay basal surface (after R.I. Zlotchevskaia et al., 1977)

The boundaries between different types of the film water layers have a kinetic nature. It means that the water molecules can diffise from adsorption water layer to the "osmosis" water layer, from the "osmosis" layers to free water, and reverse. The temperature increase intensifies diffusion of the water molecules away from the clay surface and thus causes degradation of the water films and reduction of their total splitting effect.

In spite of intensive investigation of the effect of water films on clay and colloidal stability, the quantitative theory of elastic and rheological properties of the adsorbed water layers is still not very well developed. Nevertheless, their effect on clay stability can not be ignored in formation damage evaluation.

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