

INFLUENCE OF THE IONIC STRENGTH ON THE DEPOSIT PHENOMENON AND TRANSPORT DYNAMIC OF MICROPARTICLES THROUGH SATURATED POROUS MEDIUM

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Abstract. *In this paper, the influence of ionic strength on the dynamics transport of silt micro particles through saturated sand texture is studied in the presence of repulsive interactions. The deposition phenomenon is investigated through column experimental trials. Four ionic strengths are applied by adjustment of suspension salinity (0, 5.13, 10.26 and 13.68 mM). Through this experimental study, the ionic strength influence on deposition phenomenon is shown at the micro particle scale. Ionic strength variation is the primal parameter which predicts attachment and detachment particles at constant flow. These experiments are simulated and reproduced through a numerical model based on an original deposit kinetic which is proposed in this study. This model is the coupling of two multiphase problems describing conservative salt and micro particles transport. The proposed kinetic formulations are founded on experimental tests constitutions with respect to literature trends. They are based on functional relationships between model parameters and the suspension ionic strength. In addition, the transient blocking phenomena is taken into account through a retention function Langmuir type. The suggested model shows a good match to reproduce the experimental description of the suspended particles transport under the influence of ionic strength variation. It permits to predict deposition phenomenon.*

1 INTRODUCTION

In natural subsurface and flow suspension, the micro particles can present a grateful factor to contaminant porous saturated media. Indeed, they can act as a vehicle of organic, inorganic contaminant and heavy metals or instead became a barrier to the migration of these pollutants in the case of deposition and porous medium clogging.

Chemical characteristics of the suspension flow are among the major factors involved in the process of suspended particles retention or release. Indeed, both porous texture and suspended particles are negatively charged under natural conditions. Therefore, electrostatics interactions significantly affect the physicochemical retention mechanisms. The authors ([1], [2], [3] [4], [5]) have confirmed through the investigation of experimental model system and numerical studies that increasing ionic strength enhances particles deposition rates under

constant hydrodynamic factors at typical pH values. This behavior leads to favorable attachment conditions. However, decreases in suspension ionic strength generate an increase of the electrostatic repulsion interactions and the retained particles mobilization ([6], [7]).

Nevertheless, colloidal scale transport is the major topic of these studies. The physicochemical mechanisms are dominant in the depth filtration with colloidal particles which the size is less than 1 μm [1]. Moreover, the hydrodynamic conditions influence is insignificant at this scale since the surface forces are dominant ([8], [9], [10]). In contrast, both mechanical and physicochemical mechanisms are present for particles transport with diameter between 1 and 30 μm ([11], [12]). In addition, the ionic strength influence on the micro particles suspension behavior depends also on hydrodynamic. Indeed, changes in hydro-dynamical conditions affect directly hydrodynamic forces and extension of the boundary layer diffusion mainly for micrometer particles ([13], [14]). Thus, it seems interesting to investigate the ionic strength influence on micro particles retention.

In this paper, the ionic strength influence on silt micro particles transport across saturated sand bed is studied. The micrometer silica diameter ensures the presence of both physicochemical and mechanical retention even as the size ratio of suspended particle to media grain is greater than $7.1 \cdot 10^{-3}$ [15]. To characterize the deposition phenomenon, experimental trials are conducted using a laboratory column at soil natural conditions. In these conditions, both mobile and immobile materials present negative charge surfaces at neutral pH [16]. In the first part, the followed experimental procedure is described and the used materials are exposed. In this study, particle loading of sand texture are performed at different ionic strength. In the second part, a multi-phasic model is proposed to describe micro particles transport through continuous approach. This model is the coupling of two transport phenomena of micro particles and salt. It simulates experimental tests through an original deposit kinetics proposed herein with consideration of the ionic strength variation. The applied kinetic is drew upon experiments. The model parameters are determined by reverse optimization in accord to experimental results and sensitivity analysis.

2 MATERIALS AND METHODS

2.1 Porous media and suspended particles

The porous media is composed of fine Hostun sand grain HN 31 (Sibelco, France). The sand grains have a mean diameter of 0.375 mm. They are characterized with a uniformity coefficient $C_u=1.5$ and the curvature coefficient $C_c = 1.1$. These intrinsic properties reveal a uniform distribution size and well graded porous medium. The bed sand present an hydraulic conductivity of $9.8 \cdot 10^{-4} \text{ m}\cdot\text{s}^{-1}$ and a bulk density of $2675 \text{ kg}\cdot\text{m}^{-3}$. The porosity Φ of the grain medium is equal to 0.43 ± 0.05 for each experimental trial. It is determined with an error of 0.005. The sand was thoroughly cleaned prior to use to remove organic matter and other impurities, leaving only a pure SiO_2 grain surface. The cleaning procedure consisted on the emersion of the sand in deionized water for long duration to have stabilized chemical proprieties and then it is put in oven for 24 h at 100°C .

Monodisperse suspensions of silica micro particles are used as model particles in the deposition column experiments. The particles have a mean diameter d_p between 1.3 μm and 22 μm ($d_{50}=11 \mu\text{m}$; $C_u=10.8$; $C_c = 0.948$). The microscopic description shows a crystalline

form of the silica particles with no uniform sizes. Silica particles are composed mainly of quartz SiO_2 ($> 98\%$) and a small fraction of inorganic matter (Fe_2O_3 , Al_2O_3 , K_2O , CaO , TiO_2).

The suspended particle sizes and the ratio size of particle and sand grain suggest the presence of both retention physicochemical and mechanical (straining, wedging, interception) mechanisms ([11], [15]).

2.2 Transport particles experiments

Particle transport experiments are conducted in a plexiglas column packed with clean sand grains at dry conditions and vibrated regularly to minimize layering or air entrapment. The column has an inner diameter of 7 cm and a height of 60 cm. The column dimensions are justified as they allow a homogenous flow along the sand media and reducing preferential pathways. They ensure an important pore space that approximates particles transport under natural conditions.

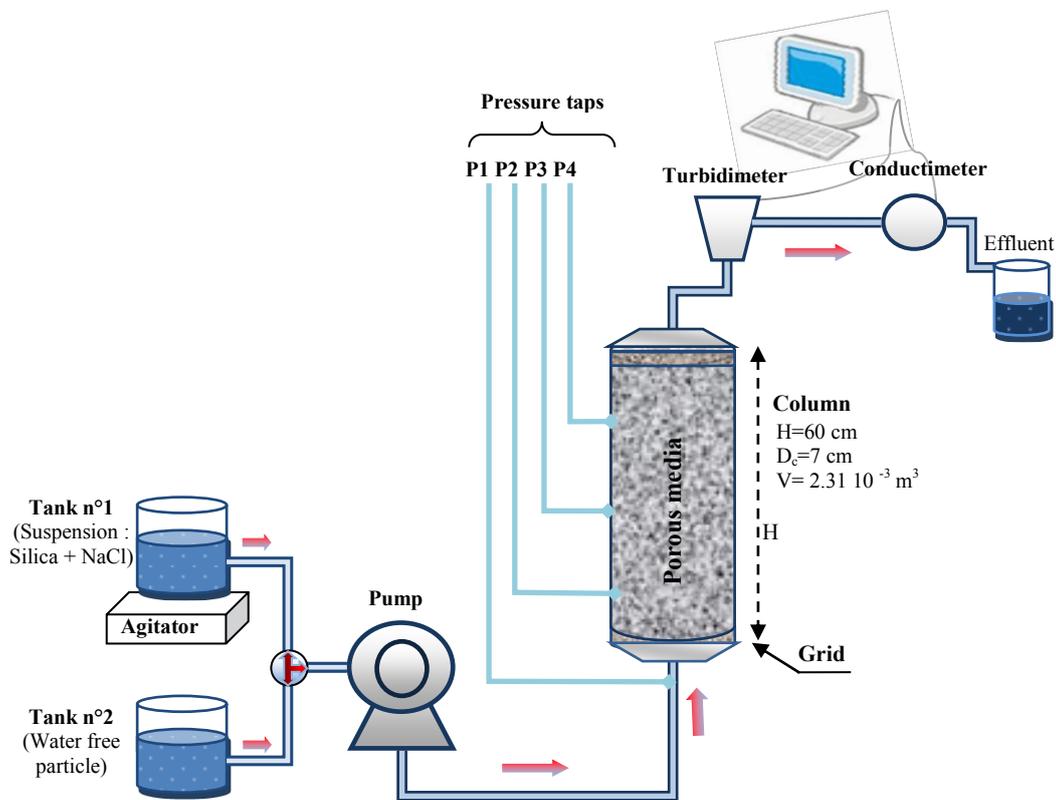


Figure 1: Experimental apparatus

A peristaltic pump (Verder AU.UPC.EZ) is connected to the sand bed. It ensures continuous injection of electrolytic solution or particles suspension through the vertical oriented column (Figure 1). The effluent properties are analyzed continuously using a turbidimeter (Turbimax CU21) and a conductimeter (Liquisys M CLM223). These measures describe respectively the particles and salt concentration of the effluent.

Four differential manometers are used to follow the evolution of the pressure at different points through experimental test in order to identify possible clogging phenomenon. The porous media has been purged with CO₂ to ensure a complete saturation. Then, it was initially equilibrated by injection of deionized water. Then, particle free background electrolytic solution is injected through the sand texture for 6 hours at the specific ionic strength used for each experimental test. This procedure eliminates traces and also impurities that can disrupt turbidity measure.

Transported particles tests are performed with a particles concentration c_{ao} of 1 g.L⁻¹ at constant darcyan velocity ($u_d=5.19 \cdot 10^{-2}$ cm.s⁻¹). A particles suspension of 3.5 V_p (pore volume) is injected for different ionic strength I (0, 5.13, 10.26 and 13.68 mM) at neutral and constant pH (6-7). The sodium chloride (NaCl) is used to adjust the ionic strength. For the pore volume injected, there is not permeability reduction through sand texture since hydraulic gradient still constant according to realized pressure measures. The particles suspension is mixed continuously to ensure the mixture homogeneity and to have isotropic properties. All the experiments are run in duplicate to verify the criterion of repeatability.

4 MODELING AND VALIDATION

4.1 The transport particles model

A continuous approach is adopted to describe the micro particles transport through the sand texture ([17], [18]). The medium is considered stable, isotropic, isothermal and homogeneously saturated. It is constituted from two phases: A solid phase (The matrix (g) and the deposited particles (p)) and a fluid phase (The interstitial fluid (w), the solute (s) and the suspended particulates (a)). The fluid phase is considered homogeneous and incompressible. The sodium chloride salt (s) is used to adjust the ionic strength I . Therefore, it is taken into account as a fluid phase component. The salt precipitation absence hypothesis is assumed.

The soil representative volume components are characterized by an absolute velocity v^α ($\alpha \in [g, p, w, s, a]$). They have an intrinsic density ρ_α relative to the constituent material (α) and a bulk density ρ^α which is defined as the ratio of the material mass m_α by the total volume V . The suspended particles and porous media grain have equal intrinsic density as assumption. In addition, the suspended particles and the salt have equal effective velocity as the intestinal fluid ($v^a = v^s = v^w$) [19].

The transport model is the coupling of two multi-phasic problems: Particle and solute transport. It is based on the synthesis of the mass conservation equations which are applied to the different medium constituents ([20], [5]). Laminar and unidirectional flow assumptions are considered. The model is presented through the following equations,

$$\phi \frac{\partial c_s}{\partial t} - \phi D_s \frac{\partial^2 c_s}{\partial z^2} + u_d \frac{\partial c_s}{\partial z} = 0 \quad (1)$$

$$\phi \frac{\partial c_a}{\partial t} - \phi D_a \frac{\partial^2 c_a}{\partial z^2} + u_d \frac{\partial c_a}{\partial z} = -R_a \quad (2)$$

$$\rho_g(1 - \phi) \frac{\partial c_p}{\partial t} = R_a \quad (3)$$

c_s is the conservative salt concentration (g.L^{-1}), c_a is the suspension particle concentration (g.L^{-1}), c_p is the deposited particle concentration, R_a deposit rate ($\text{g.L}^{-1}.\text{s}^{-1}$), ρ_g is the intrinsic density of porous medium (Kg.m^{-3}), $u_d = \phi v^w$ is the darcyan velocity (m.s^{-1}), ϕ is the porosity ($\text{m}^3.\text{m}^{-3}$), $D_s = \alpha_s u_d$ and $D_a = \alpha_a u_d$ are the hydrodynamic dispersion coefficient respectively of salt and particles ($\text{m}^2.\text{s}^{-1}$), α_s and α_a (m) are the dispersivity respectively of salt and particles.

Equation (1) represents the mass conservation and dispersion equation for salt. Equation (2) represents the mass conservation and dispersion equation for suspended particles. Then, equation (3) represents the mass conservation equation for the deposited particles. It characterizes micro particles exchange between solid and fluid phases. In the absence of clogging phenomena, the total porosity and darcyan velocity remain constant since the permeability reduction effects do not occur.

The deposition rate R_a can be expressed as a first order kinetic (4). It is expressed as a function of the deposit kinetics k_a (s^{-1}) and the retention function ψ (-) ([21], [22]).

$$R_a = \phi \psi c k_a(c_s) \quad (4)$$

The deposition kinetic k_a describes the velocity at which the suspended particles are transported from the fluid phase to the solid phase. In accord to the literature trend, suspended particles retention increases with the ionic strength. Thus, the deposition kinetics k_a may be expressed as a function of the suspension salinity. It is based on experimental results and drew upon literature considerations ([1], [23]).

$$k_a(c_s) = k_{a0} \cdot \left(1 + \frac{\kappa}{1 + \left(\frac{CDC}{c_s}\right)}\right) \quad (5)$$

κ is empirical model parameters to be adjusted with the experimental curves, k_{a0} is the deposition kinetic coefficient (s^{-1}) and CDC is the critical deposition concentration (g.L^{-1}) below which the retention phenomenon dominates. The deposition kinetic coefficient k_{a0} characterizes the suspended particles retention under favorable deposition conditions.

The retention function ψ describes the dynamic deposition phenomenon. Indeed, the deposition rate decreases progressively with the particles retention on the grains surfaces and the deposition sites saturation ([1], [24]). The retention function ψ (6) is expressed as Langmuir formulation as a function of deposited particles concentration. It involves the assumption that the deposition rate R_a depends on the particles concentration in the fluid phase even on the deposited particles fraction.

$$\psi = 1 - \frac{c_p}{c_{p,max}} \quad (6)$$

$c_{p,max}$ is the maximum concentration of deposited particles per mass unit (g.g^{-1}) at given chemical condition. $c_{p,max}$ (7) is expected to decrease as ionic strength increase [24].

$$c_{p,max} = \frac{a_1}{\rho_g} \exp(c_s) \quad (7)$$

(a_1/ρ_g) corresponds to the maximum concentration of deposited particles per mass unit under unfavorable deposition conditions with the dominance of repulsive interactions.

Model is implemented using a finite element schema that allows the resolution of equations system (1-3) and determines the evolution of resituated and retained particle

concentration. The applied 1D mesh includes 240 quadratic elements. It ensures stable numerical solution.

The boundary conditions are presented in Figure 2. The porous medium is initially considered blank, devoid of particles

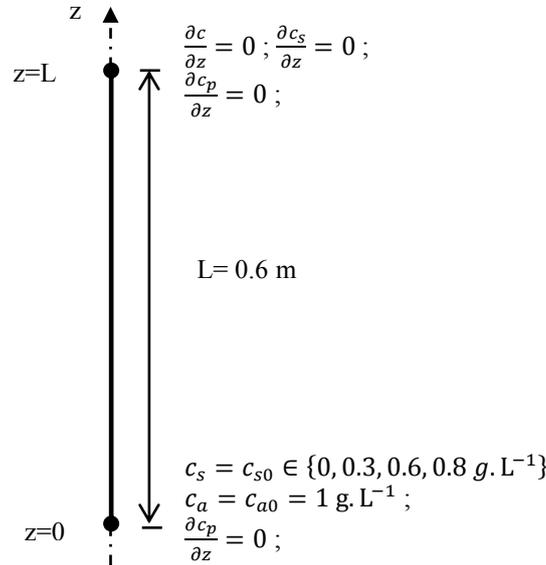


Figure 2: Model 1D of micro particle transport and boundary conditions

The model parameters identification is conducted using the successive approximations principle and a parameters sensitivity analysis. It required executing several calculations before determining the parameters in accord to experimental results. The experiments previously presented are used to adjust the model parameters. Table 1 exposes the identified parameters and their values. The micro particles dispersivity α_a is estimated equal to that of the solute α_s as simplifying assumption.

Table 1: The identified parameters values of transport particles model

Parameter	α_s (cm)	α_c (cm)	k_{a0} (s ⁻¹)	κ (-)	CDC (g.L ⁻¹)	a_1 (-)
Value	10 ⁻²	10 ⁻²	0.488	1.7	3	10.5

4.2 Sensitivity analysis

A parameter sensitivity analysis is made to analyze qualitatively the model behavior in response to a $\pm 20\%$ variation of the identified parameters change (Figure 3). The reference simulated curve is associated to experiment with ionic strength of 13.68mM ($c_{s0} = 0.8 \text{ g L}^{-1}$).

The deposition kinetic coefficient k_{a0} is the most sensitive model parameter (Figure 3.a). The k_{a0} coefficient variation ($\pm 20\%$) shows a constant and regular variation of the restitution rate (c_a/c_{a0}). However, the critical deposition concentration CDC and the model parameter κ have a moderate influence in comparison to k_{a0} (Figure 3.a-b). These parameters increase the model results accuracy.

The empirical parameter a_1 allows the increase of the model sensitivity to the salinity variations (Figure 3.c). Its influence increases over the time. Indeed, the parameter a_1 reflects the dependence of the maximum retained particles concentration c_{pmax} on the suspension salinity. It ensures the description of the particulate restitution rate increase over the time which results in the progressive retention sites saturation.

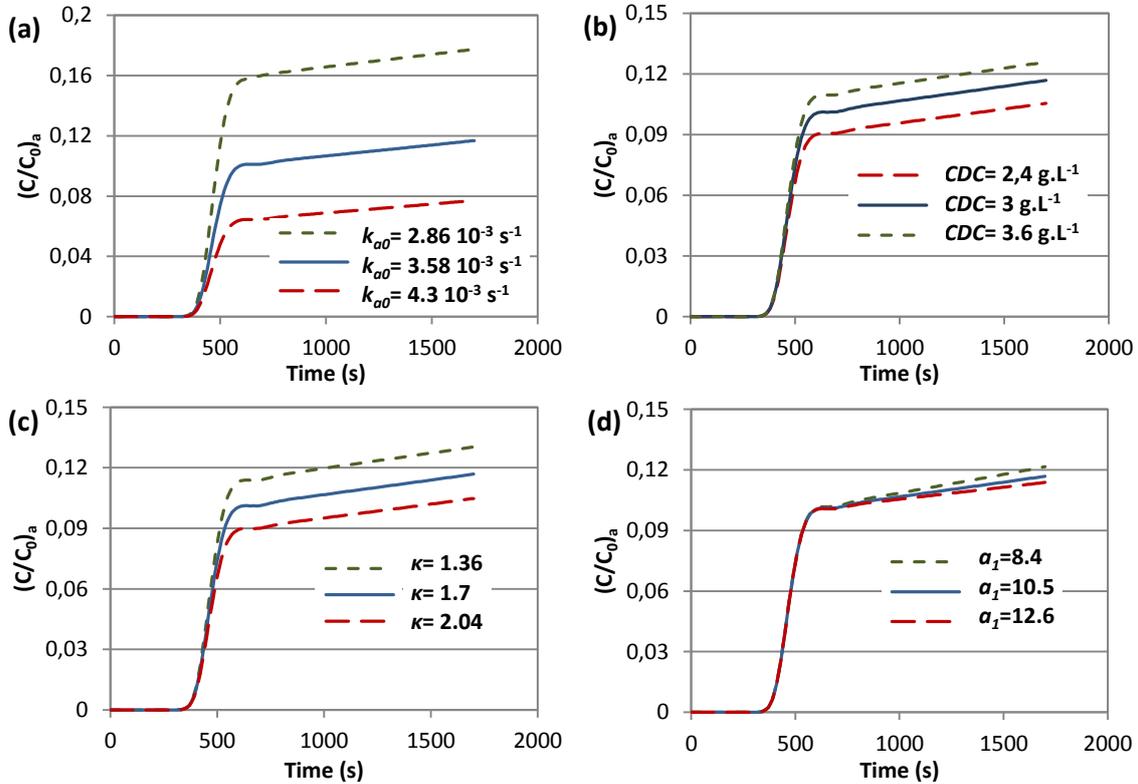


Figure 3: Sensitivity analysis of the deposition kinetics parameters (Solid line: reference curve)

4.3 Results and discussion

The particles release curves are shown in figure 4. These curves represent a typical resituated particles rate variation over the time. The experimental and numerical data are displayed as normalized resituated particles concentration (c_a/c_{a0}) at the outlet of the column as a function of the time.

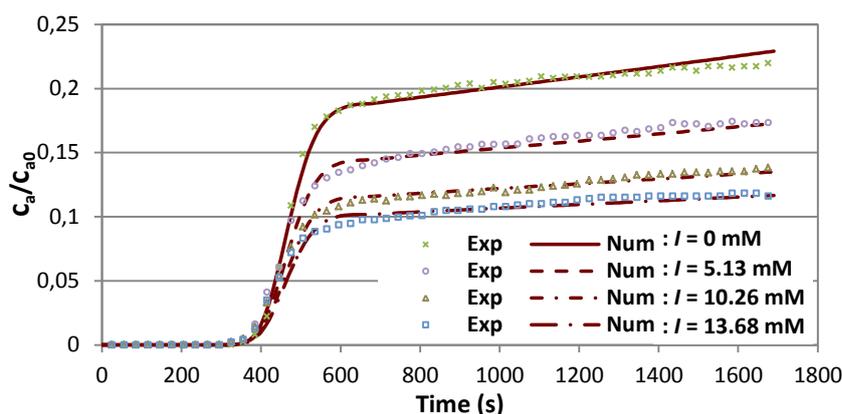


Figure 4: Evolution of the restituted suspended particles rate c_a/c_{a0} as a function of time-monotonous tests ($c_{a0}=1\text{ g L}^{-1}$; $u_d=5.19\cdot 10^{-2}\text{ cm s}^{-1}$)

c_{a0} are the silica particles injected concentration and at the inlet of the column and I is the suspension ionic strength.

The simulated experiments allow the parameters identification associated to the deposition rate R_a (Table 2). These curves show good agreement with the experimental results for the different ionic strength. In particular, the adopted deposition kinetic reproduces correctly the increase in the particles retention rate with ionic strength augmentation.

Indeed, figure 4 shows an increase in the deposition rate with ionic strength under constant flow. This result may be clarified by the classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory ([25], [26]). It describes the surface interaction between particles and porous medium grains: The system stability results in the sum of the Van der Waals attractive forces and the repulsive electrostatic forces due to the double layer. The salinity augmentation results in a reduction in double layer thickness with ionic strength increase. It is shown that the influence of electrostatic interactions which are characterized by the Debye length is inversely proportional to the ionic strength [27]. Thus, double layer thickness reduction results in repulsive forces decrease and then increasing particles deposition on the grain surface.

In addition, the model can well reproduce the quasi-linearly increasing level of particulate restitution over the time (Figure 4). This growth can be assign to the gradual decrease in physicochemical and mechanical retention sites. Indeed, the suspended particles deposit and occupy the available retention sites over time. Then, restitution particles rate is increasing at the column outlet as the deposition sites is limited. Thus, complete deposition sites saturation is possible so that restitution rate may be equal to one at long-term. But, the clogging phenomenon should eventually disrupt this trend.

Experiments have been carried out with colloidal particles in a similar framework by other authors ([24], [22], [28], [5]). The colloids present similar behavior likewise silt particles. Nevertheless, these tests show in general a rapid increase in the restitution rate ($c_a/c_{a0} = 1$) at low even high salinity. However, in accord to figure 4, obtaining an increasing level may reflects with no doubt the fact that the tests are carried out with a long column in this study. The column length implies a substantial amount of material and so more retention sites. Then saturation of all column deposition sites requires a long duration not reached in these

experiments.

5 CONCLUSIONS

This study is devoted to apprehend the suspended micro particles deposition in saturated porous media with the presence of both physicochemical and mechanical retention mechanisms. The performed experiments investigate the influence of ionic strength on the dynamics transport of silica micro particles through sand texture. In accord to performed experiments, high ionic strength promotes retention of suspended particles and leads to favorable deposition phenomena.

The experimental tests are simulated through a numerical model based on a first order deposition kinetic. This model is the coupling of two multiphasic problems. It describes the conservative salt and micro particles transport. The deposition kinetics is founded on functional relationships between model parameters and the suspension ionic strength. The proposed formulations are based on experimental results. The model shows a good match to reproduce the experimental description of the suspended particles transport through saturated medium under the ionic strength disturbances. It presents a useful tool to predict deposition phenomenon with the absence of porous media permeability reduction.

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