Experimental study of the filtration of nanofluid with SiO$_2$ nanoparticles in porous media

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Abstract. The paper considers the filtration of ethylene glycol with SiO$_2$ nanoparticles through the porous sample formed by microsphere packed bed. The experiments were performed for nanoparticles size of 7 nm, microspheres size of 150 µm, and particles volume concentration varying from 0.005 to 0.02. The data on local permeability reduction were obtained for various pressure gradients, and the peculiarities of nanoparticles retention were discussed. It was shown that nanoparticles could be transported through the porous sample, and the retention mechanism could be reversible adsorption on the pore wall. The reversible absorption mechanism of the nanoparticles retention was confirmed by the equilibrium permeability reduction determined by the magnitude of the pressure gradient.

1. Introduction
The depletion of Russia's traditional oil fields necessitates the search for methods of oil recovery from unconventional reservoirs, such as the Bazhenov suite. The collector of the Bazhenov suite is presented by kerogen–clayey–carbonate–siliceous rocks. Therefore the breakaway of the clay nanoparticles during filtration causes the formation damage and considerable reduction of its permeability. The main problems which are relevant to a fundamental understanding of deep bed filtration are the nature of and the conditions leading to the retention of particles throughout a porous sample, the change of the pore structure due to deposition, and its effect on formation performance. Typically, the following three mechanisms for retaining nanoparticles in the pores are distinguished: attachment, mechanical filtration, and sticking of particles in narrow pore throats (straining) [1]. The first retention mechanism is characteristic for nanoparticles (colloids) and it is often called physicochemical filtration. In published papers, the influence of various parameters, such as the particle size, the pore size, the ionic stress and the filtration rate on permeability decrease were studied. It was shown that the ratio of the particle size to the grain size to a large extent defines the pore plugging [2, 3]. Another important parameter is the particle content in a fluid [4]. The filtration of the nanofluid based on water and silicon oxide nanoparticles in the microspheres packed bed and sedimentary rocks was studied in [5] and [6], respectively. It was shown that surface modified nanoparticles can be transported through the sedimentary rock and the retention mechanism is the reversible adsorption on the pore surface. However, there is little literature on pore plugging by nanoparticles, and the available experimental data are still not enough for formulating an adequate model of the formation damage.
The presence of several different mechanisms of the pore plugging significantly complicates the formulation of an adequate model of porous sample damage during the filtration of liquid containing nanoparticles. In this paper, the study of physicochemical filtration with a small ratio of the size of nanoparticles to the size of grains of the porous media when mechanical confinement is not decisive is selected as the primary task. In this case, the nanoparticles are small enough to be retained in the pore throats, but nevertheless, they can produce the reservoir damage. The results of experimental studies of the filtration of ethylene glycol containing silicon oxide nanoparticles through the monodisperse packed bed of aluminosilicate microspheres are presented and the mechanism of particle retention is discussed.

2. Experimental apparatus and measurement procedure

The experiments were performed for various pressure gradients using ethylene glycol as the base liquid. The setup for study of the nanoparticles deposition in porous sample is shown in figure 1. The setup consists of porous sample 1 with the length of 0.05 m, measuring cylinder 2, differential pressure probes 3, the tank with constant level 4, measuring system 5 and web-camera 6. To determine local variation in permeability of the porous sample, the pressure drop is measured by differential pressure probes located at as shown in figure 1. The first pressure sampler is located at a distance of 2 mm from the inlet and subsequent samplers are located at a distance of 12 mm. Using the measured filtrate flow rate and pressure drops, the average and local permeability are determined. The filtrate flow rate is measured using the web-camera and measuring cylinder placed at the output of the porous sample.

The packed bed used as a porous sample is formed by aluminosilicate microspheres, screened on sieves of 140 and 160 microns. The microspheres are placed under vibration into the working section with the diameter of 1 cm and the length of 5 cm. The permeability of the porous sample $k_0$ approximately equals 16 $\mu$m$^2$. The filling of packed bed by ethylene glycol is carried out under vacuum. During experiments, the flow of ethylene glycol containing Evonik A380 silica nanoparticles with average size of about 7 nm is studied. These particles were prepared by a pyrogenic reaction. For averaged grain size of $D_g = 150$ mkm, the ratio of the particles size to the microspheres size $d_p/D_g$ equals 4.67 $\times$ 10$^{-5}$. When preparing the nanofluid, the necessary amount of silica nanoparticles is added to ethylene glycol subjected to vibration in ultrasonic bath for a long time. As a result, homogeneous solution is obtained, where precipitation does not occur for several days. The volumetric concentration of the nanoparticles in liquid is determined using tabular data on the density of ethylene glycol and amorphous silicon equals 2.2 g/cm$^3$. The rheological characteristics of the resulting suspension are
3. Determined using Brookfield rotary viscometer. It has been obtained that the nanofluid exhibits non-Newtonian behavior and its viscosity is dependent on the shear rate.

3. Experimental results

During nanofluid filtration in the porous sample, the volume of the filtrate in a measuring cylinder was measured. The experiments were performed with input volumetric concentrations of the nanoparticles $c_0$ in the range from 0.005 to 0.02. Based on Darcy law assumption, measured filtration rate and liquid viscosity, the change in permeability of the porous sample $k$ was determined as function of the pumped volume of the nanofluid. Figure 2 shows the variation in normalized permeability $k/k_0$ depending on pumped filtrate volume divided by pore volume for the filtration of nanofluid with $c_0 = 0.02$ at temperature of 21 °C. In experiments, the A380 Evonik nanoparticles with size of 7 nm, the microsphere size of 150 μm and pressure gradient of 0.26 MPa/m were used.

Permeability reduction was determined for four sections along the packed bed. The experiments confirm considerable reduction of the permeability during nanofluid filtration due to nanoparticles retention in the pore sample. There is a significant decrease in the permeability observed and it tends to an equilibrium value determined by the magnitude of the pressure gradient. With an increase in the pressure gradient, the equilibrium value of the permeability is increased. It was shown that the permeability reduction archives approximately 90% of the initial permeability at the pressure drop of 0.3 MPa. Similar behavior was observed for other initial concentrations of the nanoparticles.

When the nanofluid with velocity $v$ flows in porous sample with porosity $\varepsilon$ and permeability $k_0$, the equivalent shear rate is determined as follows: $\gamma = 4v/8k_0^{1/3}$. The dependence of the normalized equilibrium permeability of the porous sample on the shear rate for volume content of the nanoparticles of 0.005 and a temperature of 21° C is shown in figure 3. As seen, the steady-state permeability reduction becomes less at higher shear rate. Quite reasonable explanation of this phenomenon is that particles cover the surface of the microspheres and the thickness of nanoparticles layer depends on the pumped volume of nanofluid and the shear stress. This corresponds to the lining mechanism of nanoparticles retention and reversible adsorption of the particles on the pore wall.

4. Conclusions

The distinctive feature of the presented experiments is their controllability and operation with various pressure gradients that is important for prediction of the nanoparticles retention in sedimentary rocks.
Figure 3. Dependence of normalized permeability on the shear rate at $c_0 = 0.005$ and $T = 21 \, ^\circ C$.

The experiments were performed for a very small ratio of the particles size to the microspheres size $d_p/D_g$ equal to $4.67\times10^{-5}$. For this case, the sticking of particles in narrow pore throats becomes impossible. Nevertheless, the experiments confirm considerable reduction of the permeability during nanofluid filtration for the whole porous sample. The reduction of the permeability is higher near the sample inlet where the particles concentration is larger. It demonstrates that the nanoparticles can be transported through the porous sample and the retention mechanism can be reversible adsorption on the pore wall. Therefore, the filtration of a nanofluid is completely different from the filtration of a liquid with microparticles when the external cake is formed and prevents the formation damage. The retention mechanism for nanoparticles can be identified as reversible absorption due to van der Waals intermolecular forces and interaction between the particles and the microspheres surface. The nanoparticles transport to the microspheres surface is caused both by the convection and Brownian diffusion. The reversible absorption mechanism of the nanoparticles retention is confirmed by the existence of the equilibrium permeability reduction that is determined by the magnitude of the pressure gradient. It shows an increase in the probability of nanoparticle detachment when the shear rate is increased. A reasonably complete understanding of the pertinent phenomena is essential for the formulation of a comprehensive deep bed filtration theory which can be used for prediction of the filtration when breakaway of the clay nanoparticles occurs, for example in Bazhenov suite, presented by kerogen–clayey–carbonate–siliceous rocks.

Acknowledgments
This work was performed in the Kutateladze Institute of Thermophysics SB RAS and funded by the KPFI of the SB RAS Interdisciplinary Integration Studies, Project 72.3.

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