



Research article

The study of displacing ability of lignosulfonate aqueous solutions on sand packed tubes

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Abstract. This paper presents the findings of laboratory studies of rheological properties and oil displacing ability of aqueous solutions of technical grade lignosulfonate done on the sand packed tube models. The solutions containing lignosulfonate can be useful as displacement agents in development of watered reservoirs with heterogeneous porosity and permeability. When used at high concentrations, technical grade lignosulfonate can achieve selective shut-off while maintaining the reservoir pressure. The oil displacement efficiency is improved by means of redistributing the flows and selective isolation of high-permeability zones. The use of such compositions allows increasing the sweep of low-permeability reservoir zones by created pressure differential and displacing the residual oil.

Keywords: viscosity; flow curves; lignosulfonate; oil displacement ratio; sand packed tubes

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Introduction. Scientists in Russia and abroad see into the opportunities of using the compositions containing lignosulfonates as the oil displacing agents in development of watered-out reservoirs with heterogeneous porosity and permeability [1-3]. Natural reservoirs normally display macroheterogeneity [4]. The oil displacement efficiency is increased by redistributing the flows and isolating high-permeability flushed zones [5, 6]. The created pressure differential aids in increasing the sweep of low-porosity reservoir zones and displacing the residual oil [7, 8].

The most commonly used displacement agents associated with water flood sweep efficiency and improvement of oil recovery are acrylamide and sodium carbonate copolymers and sulfonated monomers and polymers based on partially hydrolyzed polyacrylamide [9, 10].

In the meantime the academic community keenly sees into the issues of using technical grade sodium lignosulfonates [11, 12], water-soluble lignin sulfonates, produced in sulfite process of wood pulp production [13, 14]. Practical and academic interest towards lignosulfonates is caused by their high surface activity. High surface activity of sodium lignosulfonates in the flow environment of oil-saturated reservoirs decreases the surface tension force at the interphase between two fluid phases and helps overcoming the resistance to the fluid flow created by capillary forces in the pore space, and also aids in recovery of oil from low-permeability interlayers that have not been swept by water flooding earlier [15, 16].

According to some estimates, more than two-thirds of the oil fields in Russia are at the third and fourth (closing) stages of development. Many of these fields have both high-permeability watered-



out zones and low-permeability oil-saturated layers that have not been swept by water flooding [17]. The use of technical grade sodium lignosulfonates at the fields with similar geological setting preceded by laboratory tests of selective water shut-off technology allows increasing the displacement ratio and the total volume of produced oil.

The paper [18] reviews the potential for the use of sodium lignosulfonate with the purpose of reducing the amount of precipitated polymers and surfactants for their subsequent penetration into the reservoir. Lignosulfonates are part of gelling agents intended for the improvement of resistance to fluid flow in the reservoir during selective water isolation workovers. Their use permits increasing the water shut-off efficiency in the reservoirs flushed in saline and fresh water even with high pressure differential [19, 20].

Lignosulfonates are known as the salts of lignin sulfonic acids, usually of sodium and potassium. Lignosulfonates have a polymeric structure, surfactant properties, and weak acidity. Technical grade lignosulfonates (LS) are economically attractive as they are a by-product of pulp cooking [9, 11]. The density of liquid LS solutions varies from 1.23 to 1.26 g/ml depending on the concentration. Lignosulfonates can also be available in solid form, as a powder of brown color. Sodium lignosulfonates typically have weak acidity (pH 4.5-5.5) [21].

Lignosulfonates are put to good use in various well in various technological activities in oil and gas industry [7, 8]. They have been successfully used in drilling mud formulations since the middle of the last century [3]. They aroused the interest of the industry as displacement agents in early 80-s abroad [9, 10]. However, in spite of many research efforts and patents secured for new technologies, this trend has not become commonly used for commercial applications [11, 12]. There is a number of patents [22, 23] and studies [24, 25] in the field of acid treatment optimization involving the addition of lignosulfonates for the reduction of the rate of dissolution of carbonates and increase of the acid treatment coverage of the reservoir, chemical properties of lignin sulfonic acids produced from sodium and potassium lignosulfonates are being studied [26, 27].

Carbonate reservoir rocks containing oil and gas normally display high degree of the properties anisotropy. The anisotropy of the properties of rock composing the reservoirs of oil fields has strong impact on porosity and permeability of rock and on the field development in general [28]. The anisotropic effect is evident during petrophysical tests as a heterogeneous permeability measured either parallel or normal to bedding [29].

When water flood displacement is used, steady-state flow of the fluid through high-permeability zones is achieved. At the same time, the interlayers with poor poroperm (porosity and permeability) properties remain unaffected and contain significant volumes of oil. This phenomenon results in increased water cut of producing wells and reduces the oil recovery [30]. In response to this problem, petroleum engineers use various technologies to divert the direction of flow in the reservoir or change the properties of individual layers [31, 32], such as, conformance control and flow profile modification. This assists the water ingress into unaffected reservoir zones [33].

To date, the efficiency of oil recovery from the reservoirs by advanced commercially adopted reservoir engineering techniques remains unsuccessful in all oil producing countries worldwide, given that consumption of petroleum refinery products continues to increase globally [34]. By some estimates the residual oil is believed to be averagely 50 to 70 % of the initial in-place oil, while the average oil recovery factor rarely reaches 40 % [35].



One of the centerpieces of oil recovery enhancement are laboratory tests supporting the oil recovery enhancement interventions, which is confirmed by a number of papers written by Russian and foreign petroleum scientists. Thus, the paper [36] states that the reservoir modeling using the cores in laboratory is one of the most reliable techniques to evaluate the reservoir behavior under the influence of various processes and phenomena, including testing various enhanced oil recovery methods. Paper [37] notes that the most reliable information concerning the poroperm and physical properties of oil reservoir rock can be obtained by the core studies. These laboratory tests include the simulation of various technologies using the cores or sand packed tubes in the natural in-situ conditions, and allow evaluating the efficiency of the technologies in question by visual demonstration.

According to some sources [38, 39], to improve the fidelity of the results obtained by enhanced oil recovery technologies simulation in laboratory, one needs to take into account the specific geological structure of the rock being investigated, therefore the selection of the reservoir models is of utmost importance.

Researches worldwide have used various approaches to selecting adequate reservoir models. Thus, papers [40, 41] describe the improved methods for determination of physical and hydrodynamic characteristics on full-size core samples (displacement efficiency and relative phase permeability) that allow increasing the reliability of results obtained in laboratory when studying the reservoirs with a complex structure. Full-size core tests allow, among other things, determining the porosity and permeability cut-off values for the reservoirs [42, 43].

Paper [44] mentions various mathematical models and calculation methods as tools that can be used for the reservoir flow simulation in addition to the cores. The tasks of flow simulation in the rock pore space typically involve high dimensionality (approx. 10^9 cells), computational domain of complex geometry, complex physical processes (multiple phases, multiple components, varying temperature, chemical reactions, etc.).

Experimental studies [45] intended for validation of the technology to recover the residual oil from heterogeneous terrigenous reservoirs used sand packed tubes of the layered-heterogeneous reservoir prepared with quartz sand. Flow tests of the designed emulsion formulation were done in laboratory conditions, and have proven its water-repelling properties.

The objective of the study is to investigate into the possibility of using aqueous lignosulfonate solutions as the base for displacement agents. To that end one needs to evaluate the rheological properties of technical grade sodium lignosulfonate solutions. Rheological characteristics are important parameters in the oil reservoir flow studies. These parameters should be determined for further studies of the oil displacing capacity of solutions, including the use of sand packed tubes and core samples.

The next step is to use the results of these laboratory studies in order to assess the efficiency of using aqueous solutions of technical grade sodium lignosulfonate as displacement agents in sand packed tubes. Then, their usefulness as the limiters of water inflow from high-permeability zones should be checked.

Materials and methods. Aqueous solution of technical grade lignosulfonate produced by Perm Pulp and Paper Mill (OOO "Prikamsky Cardboard") was chosen as the object of the research.

The properties of the solution were investigated at the first phase of the study. Dry solids concentration in the solution was determined by evaporation as 202.7 g/l. Then 10 wt.% technical grade LS solution was prepared. Based on the outcome of previous tests, this concentration displayed fast settling in the bottles containing 15 and 20 wt.% LS solution, which prevented from creating identical conditions for the experiment. The rheological tests of 5 wt.% LS solutions established that viscosity of the tested solution was only slightly different from viscosity of the water. The rheological tests were carried out on Brookfield PVS rheometer as per RD 39-0147103-329-86 guidelines and rheometer user manual [46].



The second phase was dedicated to flow tests. Pending the tests, the model of 180 g/l NaCl reservoir water, non-polar kerosene (oil model) and lignosulfonate solution were prepared. The properties of saturating fluids at 21 °C were determined earlier (Table 1).

Table 1

The properties of saturating fluids

Fluid	Viscosity, mPa·s	Density, g/cm ³
Reservoir water	1.502	1.116
Kerosene	1.490	0.7700
10 wt.% LS	5.640	1.0093

A total of four sand packed tubes were prepared. Models 1 and 2 exhibited relatively high permeability, the quartz sand particles size varying from 160 to 300 μm. Sand packed tubes 3 and 4 had relatively low permeability, the particle size varying from 63 to 160 μm. Laboratory experiments intended for investigation of the behavior in the displacement of oil from layered heterogeneous reservoirs normally

involve sand packed tubes (models) of porous media [47, 48].

The case of the sand packed tube was made of stainless steel. The grains of coarse silicate sand (more than 300 μm) were glued to the inner cylindrical surface of the tube case with epoxy to increase the roughness and significantly reduce the throughput volume of the solution pumped through the tube along its walls. The inner diameter of the tube was 20 mm, its length was 300 mm, and the estimated volume of the model including the filters was 92.67 cm³ (Fig.1, a).

The tube models were filled with sand that then was tightly compacted. Filters of blotting paper and metal wire mesh were installed at the inlet and outlet of the models to prevent the clogging

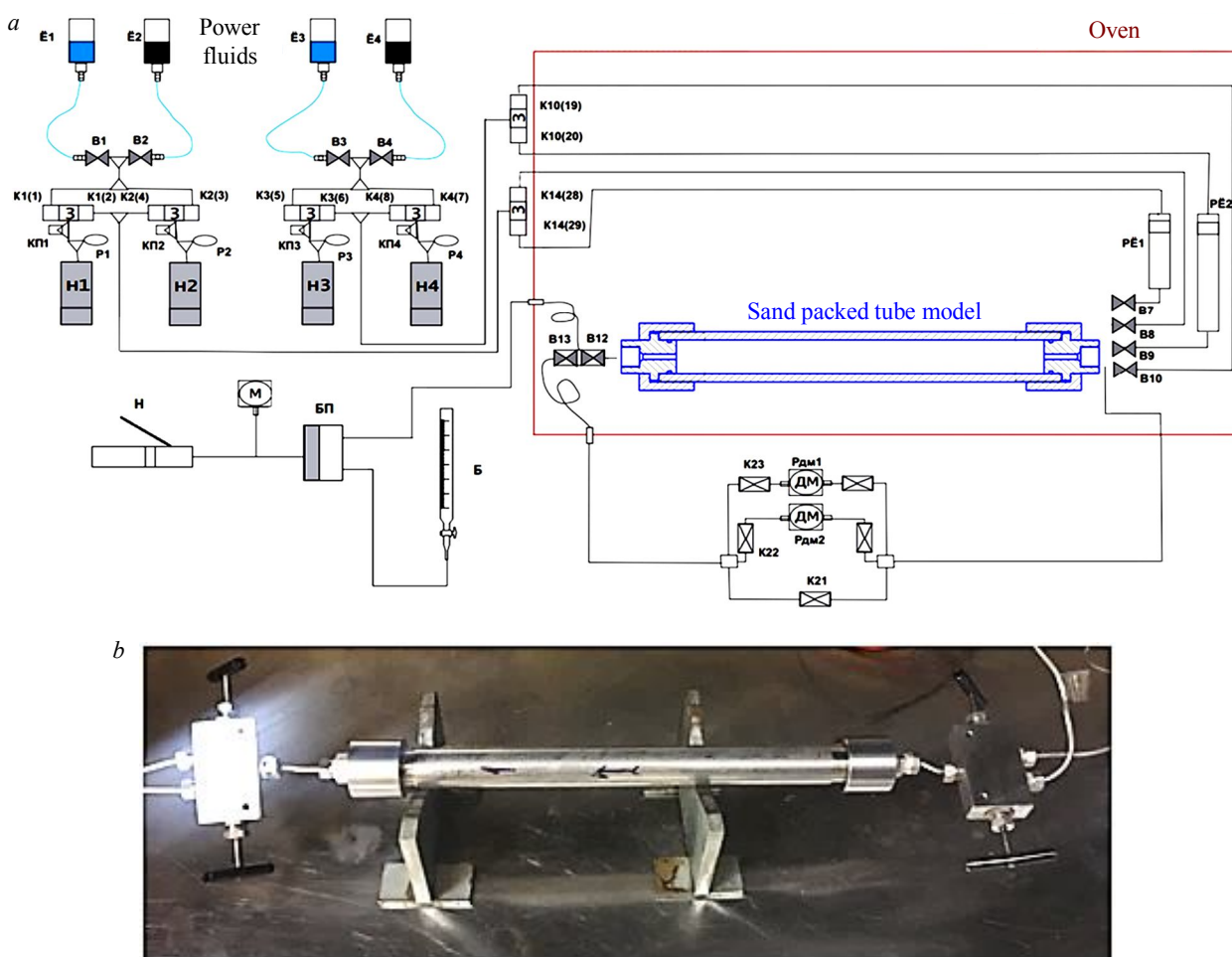


Fig.1. Schematic diagram of experimental apparatus (a) and general layout of the sand packed tube model (b)



of inlet tubes (Fig.1, *b*). The tests were done using the core analysis apparatus UIK-5(7) designed for evaluation of porosity and permeability and other parameters of reservoir rock under the pressure and temperature simulating the natural in-situ conditions.

The sand packed model prepared as describe above was put in the core analysis apparatus instead of the core holder. Constant delivery pumps created the reservoir pressure in the “piston cylinders – sand packed models system” (Fig.1, *a*). Further, saturating fluids and lignosulfonate solution were injected into the sand packed model through the piston cylinders. Backpressure was created through the backpressure unit. Pressure differential at the inlet and outlet of the model was determined by the readings of differential gauges of the test assembly. A high-precision high-pressure burette was connected to the assembly for the measurement of displaced oil volume. The simulated temperature of the sand packed tube model was 21 °C. The reservoir pressure was determined as 20 MPa.

Results. At the first stage, rheological characteristics of 10 wt.% technical grade lignosulfonate solution were investigated. The sample under investigation exhibited the properties of Newtonian fluids, i.e. the static shear stress was equal to zero and the flow curves displayed linear behavior at all tested temperatures (Fig.2).

As the temperature decreases, dynamic viscosity increases as compared to viscosity at 80 °C: 1.3 times at 60 °C; 1.9 times at 40 °C; and 2.8 times at 20 °C. The relationship of dynamic viscosity of 10 % technical grade lignosulfonate solution to the temperature is shown on Fig.3, *a*. The dynamic viscosity is linearly dependent on the concentration of technical grade lignosulfonate in the solution (Fig.3, *b*).

At the second stage of the study, the displacing ability of technical grade lignosulfonate solution was investigated.

High-permeability sand packed tubes. Once packed with sand, high-permeability sand packed tubes 1 and 2 were evaluated for their porosity, which averagely was measured at 32.72 %. The reservoir pressure and backpressure of 20 MPa was created. As the next step, reservoir water with the solids content of 180 g/l was filtered through the tubes at various flow rates. Permeability of the sand packed tubes to the modeled reservoir water was 28 and 62 D, respectively. After that,

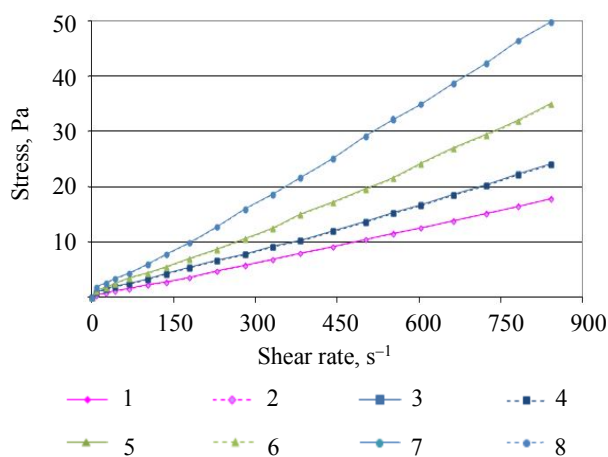


Fig.2. Rheological curves of technical grade lignosulfonate flow (10 wt.%) at various temperatures

1 – 80 °C, forward stroke; 2 – 80 °C, backward stroke; 3 – 60 °C, forward stroke; 4 – 60 °C, backward stroke; 5 – 40 °C, forward stroke; 6 – 40 °C, backward stroke; 7 – 20 °C, forward stroke; 8 – 20 °C, backward stroke

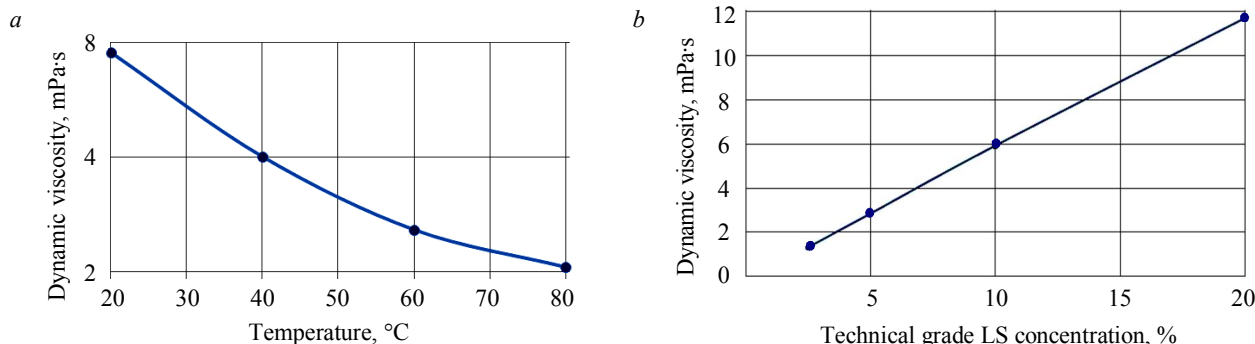


Fig.3. Relationship of technical grade lignosulfonate dynamic viscosity: *a* – at 10 wt.% concentration to temperature; *b* – at 21 °C to the concentration in aqueous solution



Fig.4. A jellylike layer of technical grade LS removed from disassembled tube

the reservoir water was displaced with non-polar kerosene, and at the same time permeability of the sand packed tubes 1 and 2 to kerosene were determined, which equaled 34 and 95 D, respectively.

At the next stage, kerosene was displaced from the sand packed tubes by the modeled reservoir water for the first sand packed tube, and by aqueous solution of technical grade lignosulfonate (10 wt.%) for the second sand packed tube (see Table 1). The displacement was done using high-precision burette. The displacement by modeled

reservoir water proceeded with constant speed, piston type displacement behavior was noted, and more than 80 % of all displaced kerosene was displaced during the flow equal to the first two pore volumes of the sand packed tube. Once the flow was stopped, the level of displaced fluid in the burette had stopped moving at the same time. The total displacement ratio was calculated in accordance with the referenced source [39] and equaled 35.80 %.

The experiment with displacement of kerosene from the sand packed tube by technical grade lignosulfonate (10 wt.%) showed a controversial result. In contrast to the displacement by modelled reservoir water, the displacement process was proceeding at varying speed, and when during the displacement by this solution the flow stopped, the fluid level in the burette continued to increase for some time. Even at the initial stages of displacement (the first two pore volumes), a water-and-kerosene mixture exited the sand packed tube, while technical grade lignosulfonate apparently blocked part of the pore channels of the sand packed tube, and was not displaced due to a significant increase in viscosity, which as a consequence notably decreased its hydrodynamic mobility. The displacement ratio was calculated as per the referenced source [39] as 60.99 %.

When disassembling the sand packed tube, it was visually established that technical grade lignosulfonate solution successfully passed through the inlet filter, and penetrated 5 to 7 cm inside the sand packed tube. During displacement, technical grade lignosulfonate formed a jellylike layer, which was due to its polymeric physical properties (Fig.4).

Low-permeability sand packed tubes. Low-permeability sand packed tubes 3 and 4 were investigated in the same way – they were filled with sand and packed similarly. After sand-packing, the model tubes were evaluated for porosity that averaged at 26.21 %. The temperature of the sand packed tube was 21 °C, and the reservoir pressure and backpressure were simulated at 20 MPa.

Then, reservoir water with the solids content of 180 g/l was filtered through the tubes at various flow rates. The permeability of model 3 to reservoir water was 315.87 mD, while for model 4 it was 358.25 mD. Such a difference was likely to be caused by varying force applied during the packing of sand into the tubes.

After that, the reservoir water was displaced with non-polar kerosene, and at the same time permeability of the sand packed tubes to kerosene was determined. Permeability to modeled oil for sand packed tube 3 was 252.52 mD, and for sand packed tube 4 it was 315.24 mD.

At the next stage, kerosene was displaced from the sand packed tubes by the modeled reservoir water for sand packed tube 3, and by aqueous solution of technical grade lignosulfonate (10 wt.%) for sand packed tube 4. High-precision burette was used. The displacement of kerosene from sand packed tube 3 by modeled reservoir water proceeded fairly evenly, as indicated by displacement behavior of non-polar kerosene. Most of the kerosene was displaced during the flow equal to the first two pore volumes of the sand packed tube. Once the flow was stopped, the level of displaced fluid in the burette had immediately stopped moving. The displacement ratio equaled 22.85 %.

The process of kerosene displacement in model 4 by technical grade lignosulfonate aqueous solution (10 wt.%) was similar in behavior to the experiment ran on model 2. In contrast to displacement by modeled reservoir water, once the flow was stopped during the displacement



by lignosulfonate solution, the level of fluid in the burette continued to rise for some time. Even at the initial stage of displacement (the first two pore volumes), a water-and-kerosene mixture exited the sand packed tube, while technical grade lignosulfonate apparently blocked the pore channels of the sand packed tube, and was not displaced. The displacement ratio was 28.95 %. When disassembling the sand packed tube, it was visually established that technical grade lignosulfonate solution passed through the inlet filter, and penetrated 5 to 7 cm inside the sand packed tube, just as it did in the previous experiments.

For the results of the tests see Table 2 below.

Table 2

Flow test results

Parameter measured	Sand packed tube 1 (160-300 μm)	Sand packed tube 2 – LS (160-300 μm)	Sand packed tube 3 (63-160 μm)	Sand packed tube 4 – LS (63-160 μm)
Permeability to reservoir water, D	28	62	0.32	0.36
Permeability to kerosene, D	34	95	0.25	0.31
Displacement ratio, %	35.80	60.99	22.85	28.95

Discussion. The results obtained through rheological tests show that in the range of temperature between 20 and 80 °C lignosulfonate solution in question exhibits the properties of Newtonian fluids. As the temperature increases, the viscosity of the solution decreases, and this relationship is exponential.

It was established that the use of technical grade lignosulfonate as a displacement agent can significantly increase the oil displacement from high-permeability sand packed tubes, and slightly improve the displacement ratio in low-permeability environment. Following the outcome of the study, have arrived at the conclusion that during filtration the particles of lignosulfonate settle in relatively large pores and channels of the reservoir model by gravity force, and improve the conformance in the pore space.

In low-permeability sand packed tubes, rock particles were likely to have almost similar grain size, and the pore space had a relatively homogeneous structure, so the particles of technical grade lignosulfonate settled in the pore space of the sand packed tube uniformly, and had a lesser effect on the displacement ratio increase.

Conclusion. The study involved the investigations of rheological properties of technical grade lignosulfonate aqueous solution (10 wt.%). The solution was investigated to be used as the base of displacement agents intended for redistribution of fluid flows in oil reservoirs. The selective water shut-off is achieved among other things by increased viscosity of the composition as compared to reservoir water, as well as by flocculating clots that isolate high-permeability reservoir zones, ensuring increased sweep efficiency from low-porosity zones of the pore space, while the surfactant properties of the solution aid in additional displacement of the residual oil. The study has established that during the displacement of modelled residual oil by technical grade lignosulfonate solution, the displacement ratio has increased averagely by 41.3 % for high-permeability sand packed tubes, and by 21.1 % for low-porosity sand packed tubes.

The obtained results show that at high concentrations technical grade sodium lignosulfonate aqueous solutions probably act as effective water inflow limiters in high-permeability heterogeneous reservoirs. For a more accurate characterization of these solutions, additional tests with the oil reservoir cores will be required.

When technical grade lignosulfonate aqueous solutions (10 wt.%) were used as a displacement agent in the sand packed tubes with the properties comparable to those of real-world oil reservoir rock, the displacement ratio had increased in a lesser degree, all other conditions being equal.



Due to the need to collect an array of data from the results of the experiments carried out in various conditions (temperature, pressure, salinity, composition of reservoir water, and rock lithology), the results are preliminary, if used for statistical processing.

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