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Microfluidic investigation of oil displacement efficiency using nanoemulsions

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This paper presents the results of a microfluidic study of the process of oil displacement from a model porous medium by a nanoemulsion. Nanoemulsions were prepared on the basis of distilled water and diesel fuel with a concentration of 1 wt.%. The mass concentration of the emulsifier varied from 0.05 to 0.4 %. A technique for preparing stable emulsions based on the method of large droplet crushing and ultrasonic treatment was developed. To study the mechanisms of the effect of emulsions, their wettability and interfacial tension characteristics were studied. As a result of the microfluidic experiments, it was found that nanoemulsions with a low content of the hydrocarbon phase make it possible to increase the oil displacement coefficient compared to water from 32 to 57 %. In this regard, further systematic study of their properties seems promising for the development of new methods for enhancing oil recovery.

Keywords: nanoemulsions, microfluidic chips, interfacial tension, wettability, oil displacement.

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The number of studies focused on the use of nanoemulsions (dispersed systems with a maximum average size of dispersed phase droplets on the order of 100 nm) in the oil industry has increased significantly in recent years. The capacity to affect significantly the wetting characteristics of formations is combined in nanoemulsions with a number of unique properties that make them much sought after in the oil industry. One of these properties is their relatively high resistance to stratification and coagulation [1,2]. Another important feature of nanoemulsions is the droplet size, which is smaller than the effective pore diameter of reservoir rocks. This facilitates the passage of the displacement fluid through the pore structure with less risk of plugging and loss of formation conductivity. According to many researchers, nanoemulsions have the greatest potential of all displacement agents for enhancing oil recovery [3,4].

The use of nanoemulsions in oil recovery enhancement is currently being studied extensively through various methods. In the present paper, we report the results of a microfluidic study of the process of oil displacement from a model porous medium by nanoemulsions. Microfluidics provides fundamentally new insights into the displacement process [5]. Such data are inaccessible to traditional research methods and make it is easier to analyze the structure and dynamics of the displacement front, examine the behavior of menisci at the point of phase contact, etc. This allows one to gain a better understanding of the mechanisms of influence of the displacement agent on capillary-held oil. The currently available amount of microfluidic data on the flow regimes of nanoemulsions during oil displacement is insufficient.

Nanoemulsions for flooding were prepared based on distilled water. Diesel fuel (Gazprom Neft PJSC, GOST 305−82) with a viscosity of 3.5 cP and a density of 830 kg/m^3 was the dispersed phase. The PC-501 emulsifier (NPO „Reagenty Sibiri") was used to make the emulsions stable. The mass concentration of the emulsifier varied from 0.05 to 0.4 %. Very low values of the hydrocarbon phase concentration were studied; the presented results correspond to a mass concentration of diesel fuel in the emulsion of 1 %. Nanoemulsions were used to displace oil with a density of 901.3 kg/m^3 and a viscosity of 79.3 mPa·s.

A technique based on the method of large droplet crushing was developed in order to prepare stable nanoemulsions. The method consists in slow (dropwise) simultaneous introduction of the hydrocarbon phase and the emulsifier under continuous mechanical stirring with a high-speed stirrer (OFITE 152-18-Prince Castle, 20 000 rpm, 60 min) with subsequent ultrasound processing („Volna-M" unit, 22 kHz, 400 W, 30 min) with temperature control. The use of this technique and adjustment of the emulsifier dosage and type made it possible to obtain stable nanoemulsions with a droplet size on the order of 100 nm.

The influence of emulsifier concentration on the efficiency of oil displacement from a microfluidic model of a porous medium was investigated. A Dolomite microfluidic chip, which allows one to model complex porous structures of rocks (Fig. 1), was used. The chip is produced by etching sodium-lime glass with hydrogen fluoride, which is followed by thermal bonding. It has one input and one output; the standard size of the microfluidic chip without connectors is 92.5×15.0 mm with a thickness of 4 mm, and the porous region of the chip is 10×60 mm in size. The length of the input channel (forks included) is 27.7 mm, and its volume is 0.9 μl. The output channel has a length of 99.2 mm with an internal volume of 3.2 μl.

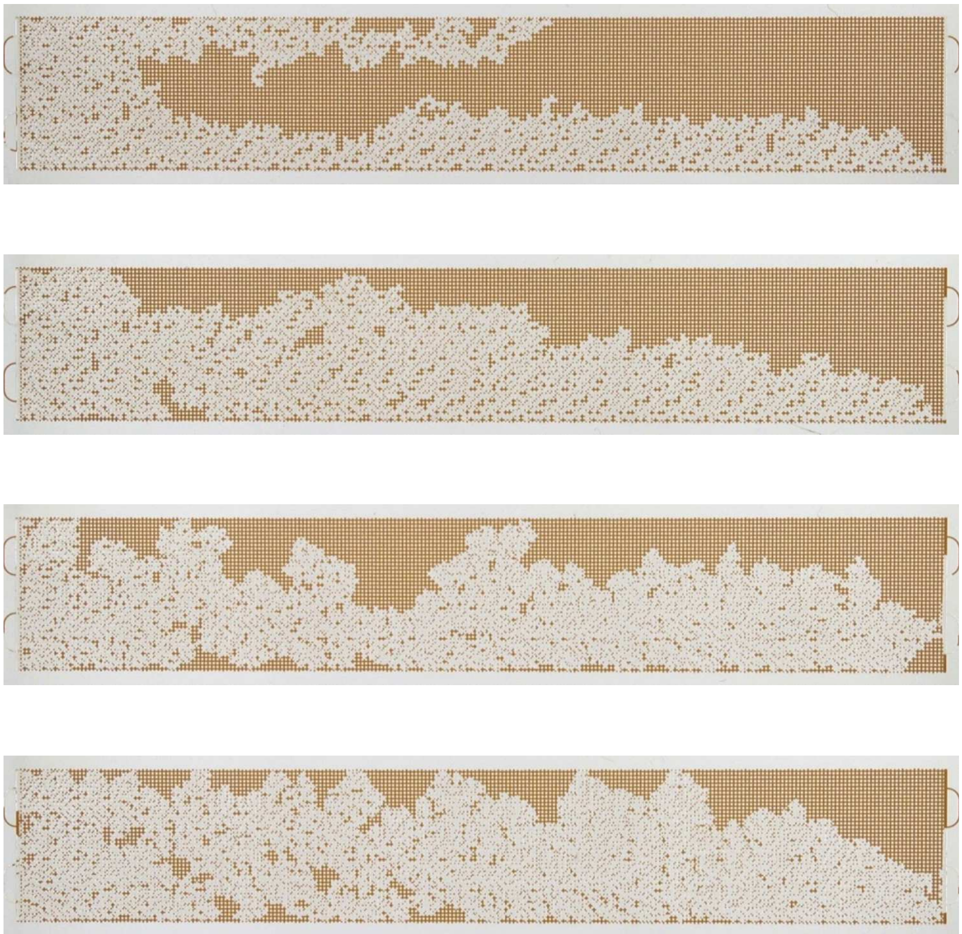
a
b
c
d

Figure 1. Photographic images of oil distribution in a microfluidic model after displacement by water (a) and nanoemulsions with an emulsifier concentration of 0.1 (b), 0.2 (c), 0.4 % (d).

The total length of the porous region is 4800 mm, and its volume is $\sim 38\,\mu$l. The surface roughness of the channels is 5 nm. Constrictions („pores“) formed in the channel network are distributed randomly to imitate the natural structure of rocks. The network contains 38 pores with Ø63 μm, 40 pores with Ø85 μm, and 50 straight channels with a nearly elliptical cross-section (channel depth and width are 100 and 110 μm, respectively).

A detailed description of the experiment was given in [6]. An SPLab02 two-channel syringe pump with its pumping rate ranging from 0.831 nl/min to 127 ml/min (± 0.5 % error) was used in experiments. The displacement fluid was supplied via a 1 ml Hamilton syringe with a Luer fitting.

The microfluidic chip was positioned horizontally on top of a glass slide. A Sony RX100 IV camera was used for photo and video recording. The empty chip was first filled completely with oil, which was then displaced by the nanoemulsion at a fixed flow rate of 0.5 μl/min. Several pore volumes were pumped to reach a steady residual oil saturation state. Figure 1 shows photographic images of the microfluidic chip at the final stage of injection of the nanoemulsion with different emulsifier concentrations. The oil displacement factor was estimated by processing the images obtained in the course of experiments using the BlackBox Component Builder application (Oberon microsystems, Switzerland) and the FreeImage library based on the HSV color model [6]. The image of the microfluidic chip consists of pixels belonging to the pore space and the background. In order to determine the fraction of oil, the number of pixels occupied by oil was calculated. The number of image pixels occupied by the displacement agent was calculated in a similar manner. The oil displacement factor was determined as the ratio of the number of pixels occupied by the displacement agent (water/emulsion/aqueous solution of a surfactant) to the number of pixels occupied by oil.

The experimental data revealed that the front of displacement by water moved very unevenly. The flow of water in the oil-filled porous medium broke up into separate streams. Water finally reached the exit of the micromodel in approximately 30 min; the process then got stabilized, and oil displacement ceased. Figure 1, a shows the residual distribution of oil in the microfluidic chip after the process of displacement by water. It can be seen that a significant

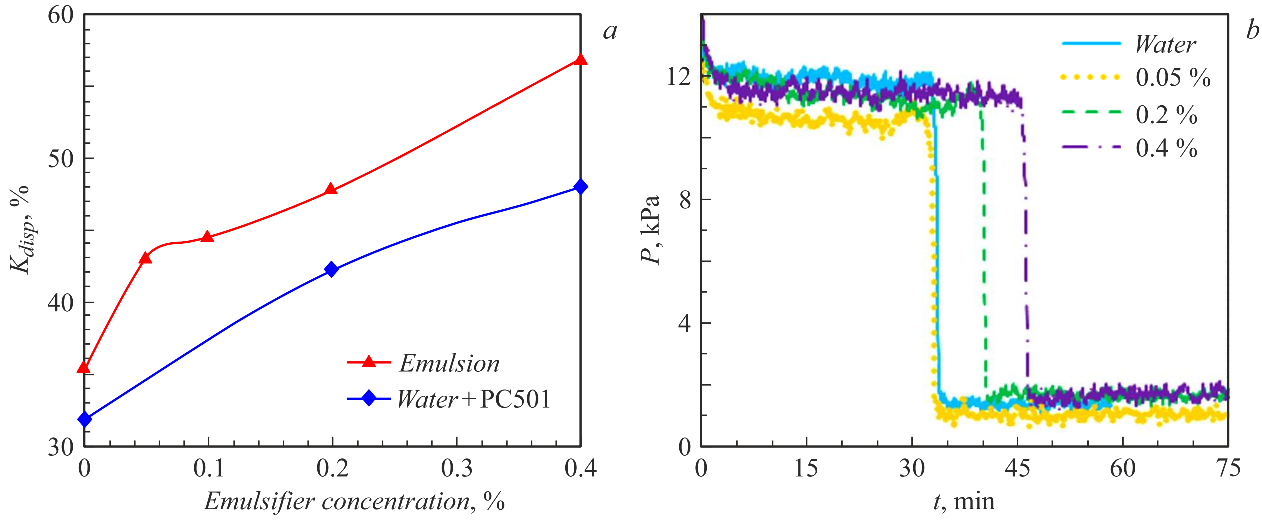

Figure 2. Dependence of the displacement factor on the emulsifier concentration (*a*) and dependence of the pressure drop on time in the process of injection (*b*).

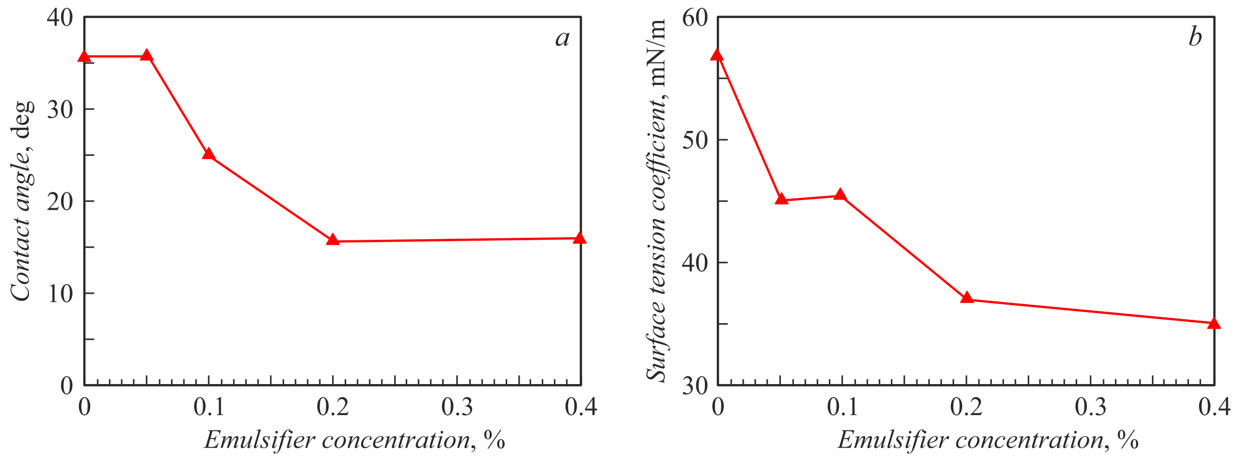

Figure 3. Dependence of the contact angle (*a*) and the surface tension coefficient (*b*) for the nanoemulsion on the emulsifier concentration.

part of the pore space remains occupied by oil; the factor of displacement by water is 32 %. When nanoemulsions were injected, the displacement pattern changed (Figs. 1, *b*–*d*). The flow was also dominated by individual jets, but the width of these jets and their tortuosity in the pore space increased significantly. Since it spread over a large part of the pore space, the length of the time interval within which the displacement fluid reached the exit from the porous medium increased significantly. This increase is evident in the plots of dependence of the pressure drop on time in the process of injection (Fig. 2, *b*), where a sharp reduction in magnitude of the pressure drop corresponds to the moment when the displacement fluid reaches the exit. An increase in jet width and coverage of the spore space area naturally leads to a significant enhancement of oil displacement. It can be seen that the islands of residual oil saturation are decreasing in size. The dependence of the oil displacement factor for emulsions with different emulsifier concentrations is shown in Fig. 2, *a*. It was demonstrated that the emulsion (even without an emulsifier) raises the displacement factor approximately by 10 % compared to water. The addition of an emulsifier has a positive effect on the displacement efficiency. At the same time, it can be seen from Fig. 2, *a* that the introduction of an emulsifier into water does not induce an oil displacement enhancement similar to the one seen in experiments with the emulsion. This is attributable to the fact that the surface tension coefficient at the emulsion/oil interface is lower than at the aqueous surfactant solution/oil interface; the effect is associated precisely with the presence of the hydrocarbon phase. As the emulsifier concentration in the emulsion reached 0.4 %, the displacement factor increased additionally to 57 %.

This is due to a reduction in droplet size in the process of dispersion with the addition of an emulsifier, which leads to a change in wetting characteristics and interfacial tension.

Figure 3 shows the dependences of the surface tension coefficient and the contact angle of a quartz glass plate with the nanoemulsion on the emulsifier concentration. These dependences were measured using the hanging and sessile drop methods.

It is evident that the wettability improves as the emulsifier concentration increases. The contact angle decreases from 35 to 16°, which contributes to better washing of oil from the channel walls. In addition, an almost twofold reduction in the surface tension coefficient was noted. This also contributes to better oil displacement. The capillary pressure reduction in the process of injection due to better wettability and a relief of interfacial tension is illustrated in Fig. 2, *b*.

Thus, it was found in microfluidic experiments that nanoemulsions with a low hydrocarbon phase content may raise considerably the oil displacement factor during flooding. In view of this, further systematic research into their properties for the development of new methods for enhancing oil recovery appears to be promising.

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Conflict of interest

The authors declare that they have no conflict of interest.

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