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Oil Displacement from a Porous Medium with the Aid of a Graphite Suspension

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Abstract—It has been found that N aqueous suspension of planar graphite nanoparticles exhibits properties of displacement fluid at the oil–water interface. Experiments with the Hele–Shaw cell showed that the process of oil displacement from the interface is not accompanied by the formation of viscous "fingers" as a result of development of instability at the oil–water interface.

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At present, most oil deposits are entering the final stage of development [1]. For this reason, the task of complete oil displacement from the bed is solved using physicochemical methods of bed processing along with its flooding [2]. There are modern notions about the mechanisms of action of oil-driving margins based on aqueous solutions of surfactants and polymers, alkali–acid media, etc. [3]. It is commonly accepted that most effective approach is that using mycobacterial slime flooding. However, a flooding agent used for this purpose must possess sufficient mobility and ultralow surface tension at the boundary with oil. In this case, the oil–water interface will be stable and the formation of viscous "fingers" due to water breakthrough via the oil layer will be suppressed.

Microemulsions represent complex dynamical structures that are highly sensitive to high-valence cations present in the oil bed. Growing temperature and pressure shift the phase equilibrium and increase the influence of capillary walls, while the motion in a porous collector with fractal geometry leads to its stochastic action on the microemulsion. As a result, the state of a microemulsion that was stable under laboratory conditions becomes unstable in a real bed [2]. In this context, the question arises as to whether is it possible to create a transition region (not microemulsion) with low surface tension at the oil–water interface that would have low sensitivity to the temperature and hardness of brine water.

The present work was aimed at studying the displacement ability of water containing additions of graphite nanoparticles at the oil–water interface.

It is known that a low surface tension at the oil–water interface is related to the formation of a structure consisting of liquid-crystalline layers of macromolecules [2]. These layers can be formed, in particular, by planar graphite nanoparticles with dimensions below 400 nm. Stability of this suspension is determined by the condition [4]

$$\Delta G_{\mathrm{mix}} = \Delta H_{\mathrm{mix}} - T\Delta S \le 0,$$

where ΔG_{mix} is the change in Gibbs' energy of the mixture, ΔH_{mix} is the change in enthalpy of the system, and ΔS is the change in entropy.

Therefore, a solvent for suspension should possess a specific surface energy (σ) close to the energy of a monolayer of graphite nanoparticles. This requirement is satisfied by aqueous ethanol solutions [5]. By varying the concentration of ethanol, it is possible to make σ close to that of planar graphite particles.

Under real conditions, liquids frequently exhibit the phenomenon of bedding (e.g., motion of oil in water). In this case, solution of the problem of possible motion of the oil–water interface depends on the coefficient of proportionality of the bulk of the reservoir and the angle of the bed slope. The shape of the oil–water interface also depends on the ratio of viscosities of oil and water. The formation of transition regions leads to a decrease in the velocity of motion on the bed bottom and increase in that at the bed roof. The difference of the velocities of motion at the inner and outer contours depends on the permeability of bed solid. Even simple schemes of oil displacement exhibit distortion of the shape of the oil–water interface with the formation of water fingers. The degree of stability is determined by the coefficient of mobility defined as

$$\lambda = \frac{k_{wo}\mu_{oil}}{k_{ow}\mu_B},$$

where k_{wo} is the permeability for water in the presence of residual oil, k_{ow} is the permeability for oil in the presence of residual bound water, μ_{oil} is the viscosity of oil, and μ_B is the viscosity of brine.

The contact front is stable provided that $\lambda < 1$, which implies that μ_B must not significantly increase. Therefore, a suspension of graphite (planar) particles must have a small surface tension σ at the oil–water interface, while viscosity μ_B and density ρ should obey the empirical relation

$$0.1 \ge \frac{\mu_B}{\sqrt{\sigma \rho d}},$$

where d is the average pore size (or capillary diameter) in the porous structure, or

$$k_{ow}\mu_B < k_{wo}\mu_{oil}.$$

Planar graphite nanoparticles were synthesized using a method described previously [6]. A polycrystalline graphite powder was heated in the Tamman furnace to 1000°C, withdrawn from the furnace, and quenched in a vessel with distilled water at room temperature (~20°C). Upon the interaction of hot polycrystalline graphite powder with water, some of the graphite precipitated, while the rest remained on the water surface in the form of a thin film. This film was collected and dissolved in 70% aqueous ethanol solution to obtain a graphite emulsion.

Analysis of the X-ray-diffraction pattern of the collected carbon material measured on a DRON-3 instrument using CuK_α radiation revealed diffraction peaks corresponding to graphite, turbostrate carbon, pure iron, and three modifications of iron oxide, including Fe_2O_3 (10*R*), Fe_2O_3 (100*M*), and γ-Fe_2O_3 (Fig. 1). The electron-microscopic examination of this carbon material on a UEMV-100K transmission electron microscope using standard techniques showed the presence of perfect graphite crystals with various thicknesses (Fig. 2).

The behavior of the water–oil interface was studied using the radial Hele–Shaw cell with geometric parameters $R_0 = 2$ mm, $R_\infty = 120$ mm, and thickness $b = 0.6$ mm [7, 8]. The synthesized planar carbon nanoparticles were used to prepare a suspension with particle sizes within 200–400 nm, which had a low surface tension ($\sigma = 43$ mN/m). The addition of this suspension to an oil phase reduced the initial viscosity by 0.7%, which led to an increase in the stability of the contact front ($\lambda < 1$).

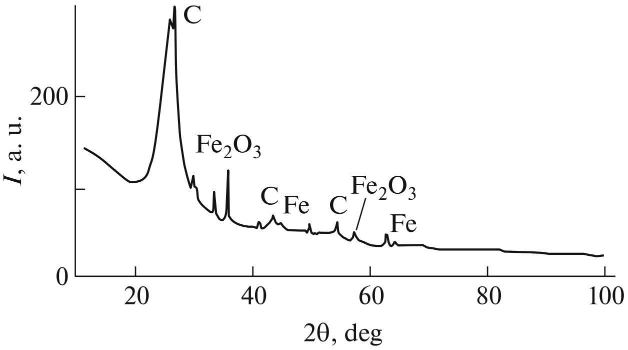

Fig. 1. X-ray-diffraction pattern of a carbon material containing planar graphite structures obtained via the interaction of distilled water with a porous carbon heated to 1000°C.

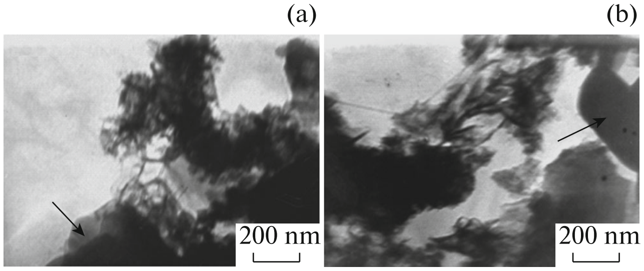

Fig. 2. Electron-microscopic images of a carbon material obtained by quenching polycrystalline graphite from 1000°C in distilled water. Arrows indicate the region containing perfect graphite crystallites (planar nanoparticles) with thicknesses (a) much below the diameter or (b) comparable with the diameter.

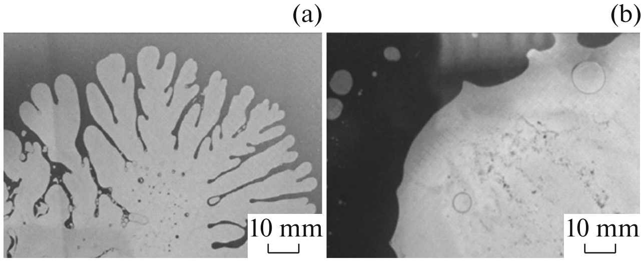

Fig. 3. Displacement of oil in the Hele–Shaw cell: (a) by pure water free of graphite nanoparticles (showing the formation of viscous fingers due to water breakthrough); (b) by water containing graphite nanoparticles (showing a stable water–oil interface without viscous fingers).

The introduction of pure water free of graphite nanoparticles into the Hele–Shaw cell with an oil phase (at a constant pressure of $p = 10$ kPa) led to the appearance of viscous fingers indicative of the instability at the water–oil interface and the water breakthrough via oil (Fig. 3a). Upon the addition of graphite nanoparticles into water, the displacement of oil proceeds with a stable front, without the formation of water fingers in the same regime at a constant pressure of $p = 10$ kPa. Figure 3b shows a clear oil–water–suspension interface.

Therefore, the proposed suspension forms a stable water–oil interface even without adding surfactants. To increase the stabilizing effect, it is possible to combine surfactants and ethanol in preparing suspensions with graphite nanoparticles so as to significantly reduce the surface tension of oil-displacing fluid.

In concluding, the results of our investigation show the good prospects of developing the technology of residual-oil displacement from gas-oil beds with the aid of graphite-based suspensions.

REFERENCES

1. *Chemical Methods in Oil Production Processes*, Ed. by N. M. Emanuel' and G. E. Zaikov (Nauka, Moscow, 1987) [in Russian].
2. *Microemulsions. Structure and Dynamics,* Ed. by S. E. Friberg and P. Bothorel (CRC, Boca Raton, FL, 1987).
3. A. Ya. Khavkin, *Hydrodynamical Foundations for the Development of Oil Deposits with Low-Permeability Reservoirs* (MO MANPO, Moscow, 2000) [in Russian].
4. I. V. Antonova, Phys. Usp. **60**, 204 (2017).
5. R. A. Soots, E. A. Yakimchuk, N. A. Nebogatikova, I. A. Kotin, and I. V. Antonova, Tech. Phys. Lett. **42**, 438 (2016).
6. E. A. Belenkov and F. K. Shabiev, Pis'ma Mater. **5**, 459 (2015).
7. L. M. Martyushev and A. I. Birzina, JETP Lett. **99**, 446 (2014).
8. L. M. Martyushev and A. I. Birzina, Tech. Phys. Lett. **34**, 213 (2008).

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