Effects of Different Power High-intensity Ultrasonic Treatment on Rheological Properties of Heavy Oil Products

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Abstract

Objectives: The objective of this study is to investigate the effects produced by intensive ultrasound on rheological properties of oil and petroleum products, especially on heavy oil products with higher concentrations. Methods: Principal method of ultrasonic treatment is represented by the exposure to the highly intensive continuous ultrasound of different power during one and the same time of treatment. The basic objects of the investigation are represented by black oil grades M-100 and M-40. The relaxation of rheological properties of the objects of the investigation was observed over the period of 10 days upon ultrasonic treatment. Findings: As a result of the study the following regularities have been identified: considerable decrease in the substance viscosity after the treatment followed by the growth in the process of relaxation; in all cases the anomalous behavior of the substance viscosity has been observed in the process of relaxation; kinematic viscosity in all experiments did not reach its initial values that used to be measured before the treatment. This means that ultrasonic treatment is capable of reducing the viscosity of heavy oil products effectively over the period of time that is sufficient for producing the heavy grades of oil and for transportation of heavy oils and petrochemicals over considerable distances. Applications/Improvements: The results of this study can be used in oil extraction and refining industries in the processes of producing and transporting the heavy grade oils and petrochemicals.

Keywords: Heavy Oil Products, High-intensity Ultrasonic Treatment, Rheological Properties

1. Introduction

Today the deposits of heavy oils account for the major share of the world reserves; however, the ratio of heavy oil in overall oil production is rather low and amounts to less than 20%. This is explained by low fluidity of this type of oils that drastically increases the production costs. Nevertheless, both absolute and relative values of the heavy oil extraction rates are growing continuously. This has been mostly predetermined by the depleted reserves of the traditional light grade oils which make it necessary to develop less technologically and economically attractive heavy oil fields. The difficulties in extracting this type of oil are mostly predetermined by its high viscosity that complicates considerably the process of pumping the oil up to the surface and its further transportation. In order to decrease the viscosity of the heavy grade oils different methods are applied primarily founded on preheating the oil carrying beds, on using solvents, and on applying reservoir fracturing techniques. However, all these methods are associated with considerable expenses and are not environmentally safe; therefore, up to now all over the world there has been a continuous search for the technologies that would be capable of reducing the oil production costs and of alleviating the detrimental impact on the environment caused by developing heavy oil deposits. One among several processes of treatment that can reduce the viscosity of these grades of oils is represented by ultrasonic treatment. This kind of treatment is practically safe from environmental perspective and it helps decrease energy consumption considerably. In this regard there is an ever growing interest in ultrasonic...
among those who strive to develop better processes for extracting heavy grade oils. These efforts have resulted in the growing number of the investigations dedicated to this subject in both oil producing and oil consuming countries. To name just a few, such developments are underway in China, Iran, Indonesia, the USA, Canada and the EU. The review of the studies dedicated to ultrasonic methods of oil extraction in China and the trends of their further development have been presented in a special publication, another review of the new ultrasonic technologies that are developed across the world has been provided in a comprehensive study. One more scientific investigation considers sonochemical approaches that have been called upon to improve the oil recovery efficiency of the oil beds. A specifically dedicated study investigated the kinetics of ultrasound effects produced on bitumen water extractions obtained from bituminous sand and also from crude oil or from residual black oil of the model contaminated soils. Iranian scientists have investigated the decrease in viscosity of residual black oil affected by the factors as follows: temperature, concentration of the solvent and the period of ultrasonic treatment. The new method of oil extraction intensification under the continuous effects of ultrasound has been described in a number of studies. Another work describes theoretical and experimental investigations of the processes of oil dehydration and demineralization affected by standing ultrasonic waves in the new reactor providing the structural parameters and the basic parameters of the reactor together with the possible modes of ultrasonic treatment. Besides, interesting information on the effects produced by highly intensive ultrasonic treatment on rheological properties of oil and petrochemicals have been described in a number of studies. It has to be noted that notwithstanding all the advantages of the ultrasonic methods, their efficiency depends considerably on the mode of application. These circumstances make it necessary that special investigations should be carried out to select the mode of ultrasonic treatment.

2. Method

In their previous work, the authors of this study have described the results of the investigations of the dependency of M-100 grade black oil viscosity on the duration of ultrasonic treatment. It has been shown that the viscosity can be reduced dozens of times and the effect can last for more than 10 days. However, in the course of relaxation the non-monotone dependency of black oil viscosity on time upon treatment has been observed that does not make for elaborating any universal mode of ultrasonic treatment.

Apart from the duration of ultrasonic treatment, another principal parameter is represented by its intensity. Therefore, there is a need to undertake the investigations of the dependency of rheological properties of heavy petrochemicals not only on the duration but also on the intensity of ultrasonic treatment.

The concrete objective of this study is to determine the effects produced by the intensity of the ultrasonic treatment on the viscosity of the oil products. Preliminary investigations have shown the direct dependency of the relative change of kinematic viscosity of the oil product after the ultrasonic treatment on its density and on the kinematic viscosity itself. The viscosity of light petrochemicals such as benzene and kerosene did not reveal any noticeable reactions to ultrasonic treatment, the viscosity of diesel fuel was affected by the ultrasound just slightly; at the same time, the viscosity of the heaviest oil products that are obtained by straight distillation of petroleum, such as black oil, were drastically affected by the ultrasonic treatment. Therefore, the principal objects of investigation are represented by the two grades of black oil that are most widely applied in Russia: M-100 black oil and M-40 black oil. M-100 grade is the product obtained by straight distillation of petroleum and it features kinematic viscosity of 14,500 – 17,000 mm²/s at a temperature of 25 °C. M-40 grade is obtained by admixing 10-12 % of diesel fuel to M-100 grade black oil and the kinematic viscosity of this product at a temperature of 25 °C amounts to 3,000 – 5,000 mm²/s.

Thereat, it should be remembered that the general objective of this study is to study the effects produced by the ultrasonic treatment on physical and chemical parameters of heavy grade oils. The principal difference between heavy and light oils is represented by higher concentration of high molecular weight hydrocarbons, i.e. the very substances that constitute such heavy oil products as black oil and diesel fuel. Therefore, the black oil was selected for the purposes of this study not as the oil product proper, but rather as a set of the chemical substances which higher concentrations make a difference between the heavy and the light oil. Consequently, these concentrations predetermine the difference in such physical and
chemical properties of heavy and light oils as viscosity, density, sound permeability, etc. The effects produced by ultrasound on these substances in their concentrated state reveal the mechanism of affecting the heavy oil properties by ultrasound.

In this study the ultrasonic treatment is represented by the effects produced by continuous ultrasonic wave of constant frequency and intensity over the period of 300 s. Continuous treatment at constant frequency makes it possible to neglect the high dissociation of the acoustic resistances that occurs between the source of the ultrasound and the loaded medium which in this case is represented by black oil grades M-100 and M-40. Due to this reason the authors of this study had to refuse using the radio-impulse and the mono-impulse types of agitating the ultrasound source, as those would require good acoustic adjustment between the source of the ultrasound and the loaded medium in a broad frequency band. If no such alignment is achieved then at the border between the two media, in this case at the border between the loaded medium and the ultrasound source there would occur great reflection of the ultrasonic waves in the process of which such high-quality ultrasound radiators as Langevin transducers prove to be the most efficient ones. Thereat, it should be noted that high-intensity ultrasonic treatment with short ultrasound impulses in many cases can be more efficient that the treatment with continuous signal. This fact has been confirmed by the existing practices of employing ultrasound in other industries. Particularly, in medical science the high-intensity impulse and radio-impulse ultrasound has been widely applied as the method of therapeutic and surgery treatment; different types of ultrasound transducers are employed, including those that use the different types of the composite piezomaterials that are acoustically aligned with the loaded medium\textsuperscript{26–28}. Also the high-intensity impulse ultrasound is used for removing the air from liquids, for removing the furring from the walls of the tubes and heating boilers and in some other technologies. The data suggested by several studies\textsuperscript{26–28} show that affecting the viscose media, including the high viscosity liquid hydrocarbons, by powerful ultrasound impulses can prove to be more efficient as compared to the effects produced by continuous ultrasound signal of the same power for destroying the intermolecular bonding interactions that are responsible for this high viscosity. However, the choice of the impulse method of ultrasonic treatment implied the availability of a great number of the parameters for the purposes of comparison, such as the amplitude of ultrasonic impulse, pulse rate, the width of the pass band, duration, etc. This would complicate the investigations very much. Therefore, at this stage of the investigation the authors of this study have selected to limit their focus by continuous ultrasonic treatment.

The ultrasound radiator was represented by Langevin transducer featuring the radiating surface of 40 mm diameter and operating at frequency of 37 kHz. The source of continuous sinusoid signal of the preset duration and power was represented by the complex that consisted of signal generator Tektronix AFG 3022 B and amplifier PA 400-5 manufactured by Precision Acoustics LTD. The signal from the generator was fed onto the amplifier and from the amplifier through the aligning transformer onto the electrodes of the radiator. The measurements of kinetic viscosity of the oil product were taken by viscometer VZ-246 featuring the operating chamber volume of 100 mm\textsuperscript{3} and the replaceable discharge flow openings of 2, 4 and 6 mm diameter. It has been calibrated applying high purity glycerin at temperatures of 22, 25, 40, 50 and 80 °C as benchmark. The relative accuracy of the measurements in the course of calibration amounted to 2.7 % with discharge flow opening diameter of 2 mm; 3% with viscometer discharge opening diameter of 4 mm and 3.5% with the opening diameter of 6 mm.

The investigations were carried out according to the methodology as follows:

- A sample was formed as 200 cm\textsuperscript{3} of the liquid under investigation in the vessel of 80 mm depth
- The temperature of the sample has been measured; to avoid temperature gradient the liquid was kept within 1 h at a temperature of 25 °C
- Applying viscometer VZ-246 the initial kinematic viscosity was measured which was necessary for its further comparing with the viscosity of the sample after ultrasonic treatment
- The radiator of the powerful ultrasonic signal was immersed into the sample to the depth of 1 mm in such a manner that the radiating surface of the transducer was in full acoustic contact with the liquid
- The transducer was fed with an uninterrupted signal of a specified power over the period of 300 s
- Upon ultrasonic treatment the temperature and kinematic viscosity of the sample were measured
The obtained results were analyzed

Steps 1-7 were repeated for the signal power values of 10, 20, 20, 40, 50 and 60 W

After the periods equal to 1, 2, 3, 4, 5, 48, 72, 96, 120, 400 hours upon the treatment the temperature and the kinematic viscosity of the samples were measured in order to trace the dynamics of the relaxation processes occurring in the samples.

3. Results

As a result of the undertaken experiments the following facts have been established:

- Affected by high-intensity ultrasound the viscosity of grade M-100 black oil was changing significantly in all cases when the electrical signals of different power were fed to the radiator according to the methodology above.
- Change of grade M-100 black oil viscosity depends considerably on the power of the fed signal.
- Kinematic viscosity of grade M-100 black oil as a result of the treatment can change several dozen times
- Affecting grade M-100 black oil by high-intensity ultrasound according to the methodology above always makes its kinematic viscosity lower.
- In the process of relaxation kinematic viscosity of grade M-100 black oil increases within the first 24 hours with all values of the ultrasonic treatment power.
- In 24 hours after the ultrasonic treatment the anomalous decrease in viscosity is observed in the process of relaxation.
- After some time (48 to 300 hours) the growth of kinematic viscosity starts again.
- In none of the cases with grade M-100 black oil did kinematic viscosity achieve its initial values after 400 hours upon treatment.

The results of affecting grade M-100 black oil by high-intensity ultrasound are shown in Figure 1. It shows the dependency of US-treated M-100 black oil kinematic viscosity reduced to 25 °C on time of relaxation within the period of 0 to 400 hours. The frequency of the ultrasound amounted to 36.6 kHz, duration of the treatment was 300 s, power rates – 20, 30 and 40 W. The initial viscosity of the batch of the black oil under investigation at 25 °C was 16,200 mm²/s. It will be observed that with low values of the intensity of treatment there are no any considerably anomalous behavior of the viscosity; however, as the intensity grows after circa 48 hours there is the anomalous growth of the viscosity followed by its decrease. This proves the importance of choosing the power of the ultrasound carefully in the process of developing the mode of ultrasonic treatment. Figure 2 shows the first 5 hours of relaxation in more detail. It will be observed that in this area the behavior of the viscosity of the black oil samples that underwent different modes of ultrasonic treatment is the same; there is the monotone growth of the viscosity with time. However, the rate of the growth is different: the viscosity value of the samples that underwent the treatment of 40 W power grows faster by far.
The results of affecting grade M-40 black oil by ultrasound are shown in Figures 3 and 4. Figure 3 shows the dependency of US-treated M-40 black oil kinematic viscosity reduced to 25 °C on time on relaxation. The frequency of the ultrasound amounted to 36.6 kHz, duration of the treatment – 300 s, power rates - 30, 50, 80 W. The initial reduced viscosity of the black oil at 25 °C was 3,100 mm²/s. Here the non-monotone character of dependency can be observed; however, this anomaly is much less vividly expressed. The first 16 hours of relaxation are shown in Figure 4 in more detail. During this period the monotone growth of the substance kinematic viscosity is observed. The rate of this growth depends on the power of the treatment and as soon as in several hours the kinematic viscosity of the samples that underwent more powerful treatment appears to be higher than the viscosity of other samples.

**Figure 1.** Dependency of US-treated M-100 black oil kinematic viscosity reduced to 25°C on time of relaxation (0 to 400 hours).

**Figure 2.** Dependency of US-treated M-100 black oil kinematic viscosity reduced to 25°C on time of relaxation, first 5 hours.

**Figure 3.** Dependency of US-treated M-40 black oil kinematic viscosity reduced to 25°C on time on relaxation (0 to 160 hours).

**Figure 4.** Dependency of US-treated M-40 black oil kinematic viscosity reduced to 25°C on time of relaxation, first 16 hours.

### 4. Discussion

The common trends of the behavior of grades M-100 and M-40 black oils when both are affected by high-intensity ultrasound and then go through relaxation after this treatment should be noted. In both cases considerable decrease in the substance viscosity was observed upon the treatment followed by its growth in the process of relaxation. Also in both cases the anomalous behavior of viscosity in the process of relaxation was observed, and in all cases kinematic viscosity did not achieve its initial values that have been fixed before the ultrasonic treatment. This means first that the obtained results are sufficiently reliable and second that there are common regularities in the changes of rheological properties of high molecular weight hydrocarbons that underwent ultrasonic treat-
ment. Drastic decrease in the black oil viscosity upon ultrasonic treatment seems to be related to the fact that powerful ultrasound destroys the intermolecular bonding interactions. Gradual growth of kinematic viscosity of the substances in the process of their relaxation is associated with the reconstruction of these interconnections. The anomalous decrease in viscosity at a certain stage of relaxation is supposedly related to the formation of the clots that have been detected in the course of taking measurements with viscometer. The most viscose fractions of the black oil were concentrated in those clots and thus, the media beyond these clots contained the less viscose fractions of the substance which predetermined the overall temporal decrease in kinematic viscosity of the substance. In the process of relaxation, the clots were growing gradually, were occupying ever greater volume and joined each other. This resulted in the renewed growth of the viscosity. The analysis of the works belonging to other authors showed that the results of their investigations of the changing viscosity of heavy grade oils and petrochemical products partially coincide with the results of this study. Practically all of the works confirm the decrease in the viscosity of heavy oils and petrochemicals affected by high-intensity ultrasonic treatment followed by the growth of their viscosity in the process of relaxation. However, the authors of this study failed to discover any statements on the anomalous behavior of the viscosity of either black oil or any other high viscosity oils or petrochemical products in the process of relaxation upon ultrasonic treatment. It may be that this only happened because the objects of the relevant investigations were different.

5. Conclusion

Intensive ultrasonic treatment decreases the viscosity of grades M-100 and M-40 black oils several times within the wide range of the values of the power of the signal that is fed to the ultrasound source. Thereat, the value of the viscosity decrease and the character of its relaxation upon treatment strongly depend on the intensity of the ultrasound radiation. Similar to the case of the ultrasonic treatment with constant power and varied durations, the anomalous non-monotone process of the black oil viscosity change is observed in the process of relaxation. This means that employing these modes of ultrasonic treatment of heavy grade oils and petrochemical products can considerably facilitate the processes of oil extraction and oil product transportation; however, each of the cases would require that the mode should be selected individually. The results of this study can be used in oil extraction and refinery industries, and also for the purposes of transporting oil and petrochemical products.

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7. References

1. Mullakaev MS, Abramov VO, Abramova AV. Development of ultrasonic equipment and technology for well stimulation and enhanced oil recovery. Journal of Petroleum Science and Engineering. 2015; 125:201–8.
2. Goland V, Kushkuley L, Mimran S, Zadok Y, Ben-Ezra S, Shalgi A, Rybianets A. P1B-7 experimental and theoretical study of strongly focused high-intensity ultrasound. IEEE Ultrasonic Symposium Proceedings, Israel; 2007. p.1305–8.
3. Fakhruddinov RZ, Ganieva TF. Petroleum processing and petroleum chemistry. Scientific and Technical Achievements and Advanced Practices. 2012; 4:10–12.
4. Khismiev RR, Petrov SM, Yu BN. Modern state and potential of extra-heavy crude oil and natural bitumen. Bulletin of Kazan Technological University. 2014; 17(21):312–15.
5. Nikitin VS, Yagodov GN, Nenartovich TL, Kuznetsov NP, Muzipov HN. Technology of oil recovery rise after super-power ultrasound impact. Oilfield Engineering. 2010; 8:14–17.
6. Mezikov VK. Prospects of applying powerful ultrasound for extra-heavy crude oil and bitumen extraction. Drilling and Oil. 2015; 7–8:72–73.
7. Mullakaev MS, Volkova GI, Gradov O M. Study of ultrasound impact on viscosity and temperature properties of oil grades having different component composition. Theoretical Foundations of Chemical Technology. 2015, 49, 3, 302-311.
8. Volkova GI, Prozorova IV, Anufriev RV, Yudina NV, Mullakaev MS, Abramov VO. Ultrasonic processing of oil to improve its viscosity and temperature properties. Petroleum processing and petroleum chemistry. Scientific and Technical Achievements and Advanced Practices. 2012; 2:3–6.
9. Wang Z, Xu Y, Suman B. Research status and development trend of ultrasonic oil production technique in China. Ultrason. Sonochem. 2015; 26:1–8. DOI: 10.1016/j.ultrasochn.2015.01.014.
10. Castañeda LC, Muñoz AD, Ancheyta J. Current situation of emerging technologies for upgrading heavy oils. Catalysis Today. 2014;220–2, 248–73. DOI: 10.1016/j.cattod.2013.05.016.

11. Abramov VO, Abramova AV, Bayazitov VM, Altunina LK, Gerasin AS, Pashin DM, Mason TJ. Sonocatalytic approaches to enhanced oil recovery. Ultrasonics Sonochemistry. 2015; 25:76–81. DOI: 10.1016/j.ultsonch.2014.08.014.

12. Abramov OV, Abramov VO, Myasnikov SK, Mullakaev MS. Extraction of bitumen, crude oil and its products from tar sand and contaminated sandy soil under effect of ultrasound. Ultrasonics Sonochemistry. 2009; 16(3):408–16. DOI: 10.1016/j.ultsonch.2008.10.002.

13. Doust AM, Eahimi M, Feyzi M. Effects of solvent addition and ultrasound waves on viscosity reduction of residue fuel oil. Chemical Engineering and Processing. Process Intensify. 2015; 95:353–61. DOI: 10.1016/j.sepproc.2015.07.014.

14. Abramov VO, Mullakaev MS, Abramova AV, Esipov IB, Mason YJ. Ultrasonic technology for enhanced oil recovery from failing oil wells and the equipment for its implementation. Ultrasonics Sonochemistry. 2013; 20(5):1289–95. DOI: 10.1016/j.ultsonch.2013.03.004.

15. Check GE, Mowla D. Theoretical and experimental investigation of desalting and dehydration of crude oil by assistance of ultrasonic irradiation. Ultrasonics Sonochemistry. 2013; 20(1):378–85. DOI: 10.1016/j.ultsonch.2012.06.007.

16. Hamidi H, Mohammadian E, Asadullah M, Azdarpour A, Eafati E. Effect of ultrasound radiation duration on emulsification and de-emulsification of paraffin oil and surfactant solution/brine using Hele'shaw models. Ultrasonics Sonochemistry. 2015; 26:428–36. DOI: 10.1016/j.ultsonch.2015.01.009.

17. Hamidi H, Mohammadian E, Junin E, Eafati E, Manan M, Azdarpour A, Junid M. A technique for evaluating the oil/hydrocarbon viscosity changes under ultrasound in a simulated porous medium. Ultrasonics. 2014; 54(2):655–62. DOI: 10.1016/j.ultras.2013.09.006.

18. Ji G, Zhou C, Zhou G. Ultrasound enhanced gradient elution of super heavy oil from weathered soils using TX100/SBDS mixed salt micellar solutions. Ultrasonics Sonochemistry. 2011; 18(2):506–12. DOI: 10.1016/j.ultsonch.2010.08.014.

19. Mohammadian E, Junin E, Eahmani O, Idris AK. Ultrasound enhanced gradient elution of super heavy oil from weathered soils using TX100/SBDS mixed salt micellar solutions. Ultrasonics. 2013; 53(2):607–14. DOI: 10.1016/j.ultras.2012.10.006.

20. Mohsin M, Meribout M. Ultrasound enhanced gradient elution of super heavy oil from weathered soils using TX100/SBDS mixed salt micellar solutions. Ultrasonics Sonochemistry. 2015; 23:413–23. DOI: 10.1016/j.ultsonch.2014.08.007.

21. Sad CM, Santana IL, Morigaki MK, Medeiros EF, Castro EV, Santos MF, Filgueiras PE. New methodology for heavy oil desalination. Fuel (Guildford). 2015; 150:705–10. DOI: 10.1016/j.fuel.2015.02.064.

22. Makarev DI, Rybyanets AN, Sukhorukov VL. Anomalous viscosity of high-molecular petroleum fractions in process of relaxation after high-intensity ultrasonic treatment. Indian Journal of Science and Technology. 2016; 9(29). DOI: 10.17485/ijst/2016/v9i29/99457.

23. Makarev DI, Rybyanets AN, Mayak GM. The possibility of creating digital piezomaterials based on piezoceramic-polymer mixed composites. Technical Physics Letters. 2015; 41(4):317–19. DOI: 10.1134/S1063785015040124.

24. Rybyanets AN, Konstantinov GM, Naumenko AA, Shvetsova NA, MAKAREV DI, LUGOVAYA MA, Elastic, dielectric, and piezoelectric properties of ceramic lead zirconatetitanate/α-Al2O3 composites. Physics of the Solid State. 2015; 57(3):527–30. DOI: 10.1134/S1063783415030270.

25. Naumenko AA, Shcherbinin SA, Makarev DI, Rybyanets AN. Novel approach to optimization of finite element models of lossy piezoelectric elements. Physics Procedia. 2015; 70:171–4. DOI: 10.1016/j.phpro.2015.08.100.

26. Rybyanets AN, Naumenko AA, Konstantinov GM, Shvetsova NA, Lugovaya MA. Elastic, dielectric, and piezoelectric properties of ceramic lead zirconatetitanate/α-Al2O3 composites. Physics of the Solid State. 2015; 57(3):558–62. DOI: 10.1134/S1063783415030269.

27. Rybyanets AN. Properties of PZT/PZT ceramic piezocomposites. Bulletin of the Russian Academy of Sciences: Physics. 2010; 75(10):1100–3. DOI: 10.3103/S1062873810080186.