Ultrasonics Sonochemistry 25 (2015) 76–81

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

journal homepage: www.elsevier.com/locate/ultson

Sonochemical approaches to enhanced oil recovery

Vladimir O. Abramov [a], Anna V. Abramova [a,*], Vadim M. Bayazitov [a], Lyubov K. Altunina [b], Artyom S. Gerasin [c], Dmitriy M. Pashin [d], Timothy J. Mason [e]

[a] Institute of General and Inorganic Chemistry of the Russian Academy of Sciences, Leninskiy Prospekt 31, Moscow 119991, Russian Federation
[b] Institute of Petroleum Chemistry of the Siberian Branch of the Russian Academy of Sciences, Akademicheskaya Avenue 4, Tomsk 634021, Russian Federation
[c] CUT-Service Ltd, Office 36, Vavilova Street 97, Moscow 117335, Russian Federation
[d] Centre of Nanotechnologies of the Republic of Tatarstan, Peterburgskaya Avenue 50, Kazan 420107, Russian Federation
[e] Sonochemistry Centre, Faculty of Health and Life Sciences, Coventry University, CV1 5FB Coventry, UK

ARTICLE INFO

Article history:
Received 7 August 2014
Accepted 13 August 2014
Available online 29 August 2014

Keywords:
Ultrasound
Enhanced oil recovery EOR
Oil well treatment
Horizontal well

ABSTRACT

Oil production from wells reduces with time and the well becomes uneconomic unless enhanced oil recovery (EOR) methods are applied. There are a number of methods currently available and each has specific advantages and disadvantages depending on conditions. Currently there is a big demand for new or improved technologies in this field, the hope is that these might also be applicable to wells which have already been the subject of EOR. The sonochemical method of EOR is one of the most promising methods and is important in that it can also be applied for the treatment of horizontal wells. The present article reports the theoretical background of the developed sonochemical technology for EOR in horizontal wells; describes the requirements to the equipment needed to embody the technology. The results of the first field tests of the technology are reported.

1. Introduction

Currently the efficiency of oil recovery from wells is less than 40% and as such is not really satisfactory [1]. Existing technologies for enhanced oil recovery (EOR) are often energy and labor intensive and often not environmentally friendly [2–6]. Thus there is great interest in the modification of established techniques and the development of new technologies. This is particularly important in the case of the newer types of horizontal wells that are generally higher yielding than traditional (vertical) types in that they harvest oil from a number of underground sources through which they pass. In Western Siberia and the Volga region one of the main approaches to the rejuvenation of failing oil wells has been through the use of chemical treatment which is useful for removing blockages. While this method is good for traditional wells it has been shown to be inefficient in the case of horizontal wells. This is because the chemical reagents injected through the wellhead often do not reach the zones along the horizontal part which need to be treated [7].

Over the last few years there has been a developing interest in physical EOR techniques, especially those based on ultrasonic treatment [8–10]. We have developed a method for EOR which includes ultrasonic treatment in the wellbore perforation zone with the simultaneous creation of a zone of lower pressure in that zone [11–12]. The methodology is particularly useful for older wells which are in the later stages of reduced yields.

Laboratory and field tests have shown that acoustical oscillations initiate a variety of chemical and physical processes in oil bearing formations. The most attractive and studied of these processes, which will be described further in this article, are:

1. Destruction of physical bonding on the boundary layer between the pores of the rock and the fluid, which hinders the movement of fluid.
2. Alteration of the fluid rheology by the destruction of the bonding between molecules in the fluid which is important in the cases of viscous and heavy oils allowing the more solid components like resin, paraffin and asphaltene to become mobile.
3. The break-down of mineral salt deposits and deparaffinization.

2. Theoretical background and laboratory scale experiments

The main difference between oil and many other viscous liquids is that its molecules form conglomerates, which account for the

* Corresponding author. Tel.: + 7 9163394568; fax: +7 4959554838.
E-mail addresses: novita@mail.ru (V.O. Abramov), anna_v_abramova@mail.ru (A.V. Abramova), vadim.bayazitov@gmail.com (V.M. Bayazitov), alk@ipc.tsc.ru (L.K. Altunina), doctrina_petroleum@mail.ru (A.S. Gerasin), dmitry.pashin@nanort.ru (D.M. Pashin), apx077@coventry.ac.uk (T.J. Mason).

http://dx.doi.org/10.1016/j.ultsonch.2014.08.014
1350-4177/

higher viscosity of oil. In these conglomerates the molecules are bonded to each other by intermolecular forces. The goal of ultrasonic treatment is to destroy these bonds and to bring the properties of the oil closer to what they would have been if no conglomerates were present.

The description of this process is in many aspects similar to the description of destruction of materials, where the kinetics of link opening depends on the temperature of the material T, the energy of bond breaking E_0 without external influence etc. The main formula of the theory of material destruction, which describes the destruction time of one bond τ_p, can be generalized for the case of intermolecular interaction [13]:

$$\tau_p = \tau_0 \exp\left\{\frac{E_0 - \gamma(\sigma_c + \sigma_u)}{kT}\right\} \tag{1}$$

In the above Eq. (1) τ_0 is a constant value, which depends on the material properties, σ_c is the static stress, σ_u is the stress produced by ultrasonic treatment, γ characterizes the degree of transmission of the average stress to one bond and is structure dependent and k is the Boltzmann constant.

σ_u is determined (Eq. (2)) taking into account the periodic nature of this stress $\sigma(t)$:

$$\sigma_u = \frac{1}{\tau}\int_0^{\tau_p} dt\sqrt{\sigma^2(t)} \equiv \frac{1}{\tau}\int_0^{\tau_p} dt|\sigma(t)| \tag{2}$$

where $\sigma(t) = \sigma_0 \sin\omega t$ and $\tau = 2\pi/\omega$. Thus Eq. (1) is transformed into Eq. (3):

$$\tau_p = \tau_0 \exp\left\{\frac{E_0 - \gamma\left(\sigma_c + \frac{2\eta N_p \sigma_0}{\pi}\right)}{kT}\right\} \tag{3}$$

where $N_p = 1/(2\pi) \times \omega\tau_p$ is the number of stress cycles needed to destroy the intermolecular bond. η characterizes the percentage of energy of the ultrasonic treatment, which goes to bond destruction.

Eq. (3) is an equation for N_p, which can be changed to

$$N_p = \pi\frac{E_0 - \gamma\sigma_c}{2\gamma\eta\sigma_0} + \frac{kT\pi}{2\gamma\eta\sigma_0}\ln\left(\frac{\omega}{\pi^2}\tau_0\frac{\eta\sigma_0\gamma}{(E_0 - \gamma\sigma_c)}\right) \tag{4}$$

In order to estimate N_p, which obviously determines the time needed to destroy the conglomerates of the molecules, one needs to know the parameters E_0 and γ, which characterize the type of the intermolecular bond. A rough estimation of N_p can be done if we use the approximate equality $E_0 \approx \gamma\,\sigma_T$, where σ_T is the stress of bond destruction. Assuming $\sigma_c = 0$ (which is the typical case), Eq. (4) simplifies to Eq. (5):

$$\begin{aligned} N_p &= \pi\frac{\sigma_T}{2\eta\sigma_0} - \frac{kT\pi\sigma_T}{2E_0\eta\sigma_0}\ln\left(\frac{\pi^2}{\omega\tau_0}\frac{\sigma_T}{\eta\sigma_0}\right) \\ \tau_p &= \pi^2\frac{\sigma_T}{\eta\sigma_0\omega} - \frac{kT\pi^2\sigma_T}{E_0\eta\sigma_0\omega}\ln\left(\frac{\pi^2}{\omega\tau_0}\frac{\sigma_T}{\eta\sigma_0}\right) \end{aligned} \tag{5}$$

The energy of the intermolecular bond is related to the stress of the bond destruction is given by Eq. (6):

$$E_0 = \pi R_c^2 R_{cr}\sigma_T \tag{6}$$

where R_c is the typical "radius" of one molecule, which is about 0.5 nm for oil and R_{cr} is the critical distance required to break the connection, which was considered to be 0.5 nm (the size of molecule) for the estimation.

The stress produced by the acoustical field is equal to the acoustical pressure (Eq. (7)):

$$\sigma_0 = 2\pi f\rho cA\sqrt{\frac{\pi}{\omega}} \tag{7}$$

where f is the frequency of ultrasound, A is the amplitude of the signal, c is the speed of sound and ρ is the density of the media.

In the case of low temperatures the second quantity is low; the temperature rise due to ultrasonic treatment can be neglected as ultrasonic treatment is usually accompanied by pumping out the well by a jet pump, thus creating a continuous flow.

Thus the treatment time may be roughly estimated using Eq. (8):

$$\tau_p = \frac{E_0}{2R_c^2 R_{cr}\eta\sqrt{\pi\omega}f\rho cA} \tag{8}$$

The following numerical values were used for the estimation: f = 20,000 Hz, $E_0 = 1.7 \times 10^{-20}$ J (typical value for non polarized molecules), ρ = 900 kg/m^3 (for oil), c = 620 m/s, η = 0.3 and A near the downhole tool was determined experimentally and was equal 2 μm. In order to calculate the amplitude of the pressure depending on the distance from the acoustical emitter r Eq. (9) was used:

$$A_r = Ae^{-\alpha r}, \tag{9}$$

The damping coefficient is dependent on the frequency of the signal (Eq. (10)):

$$\alpha = \omega/c \times 1/Q, \tag{10}$$

Q is the quality factor. For materials of the earth's crust Q is usually about 300. However it should be taken into account that in case of oil formation we deal with a porous media, thus the signal is damped more. As recommended in Ref. [14] for this case we assumed that the quality factor is equal 30. Thus in our case the damping coefficient α may vary from 4 up to 5.2 depending on the structure of the porous media.

Based on the assumptions described above we have been able to calculate the treatment time needed to destroy the intermolecular connections. Near the downhole instrument it is 30 s. However depending on the structure of the porous media this time reaches 20–55 min for zones which are 1 m away from the tool (based on experiments in a barochamber it was determined that the acoustical signal is damped almost completely within 1 m of formation, thus the treatment time was calculated for this case).

The theoretical model described above was tested experimentally by studying the effect of ultrasound on the viscosity of oil. A schematic of the equipment used is shown on Fig. 1. We have used the generator TS4M1, the waveguide system had an operating frequency 20 kHz with an emitting surface 6.6 cm^2.

We have studied the viscosity changes of oil after 3 min of treatment. The power of the generator was 3 kW. In order to avoid the

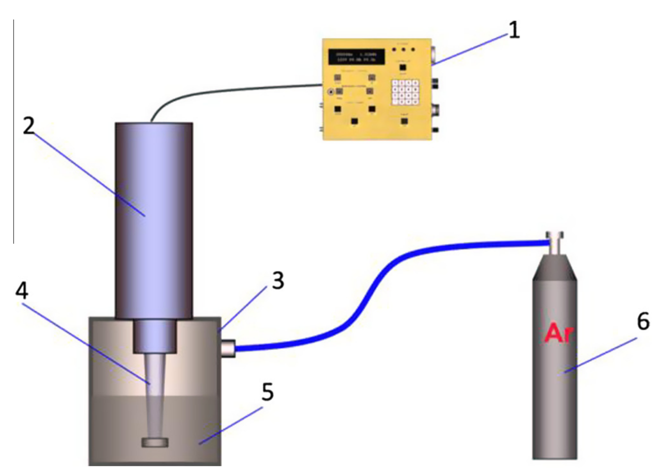


Fig. 1. Equipment scheme for studying the effect of ultrasound on the viscosity of oil: 1 – ultrasonic generator, 2 – magnetostrictive transducer, 3 – sealed reactor for treatment of oil, 4 – waveguide system, 5 – oil, 6 – compressed gas cylinder to maintain the pressure in the reactor.

Table 1
Characteristics of oil from the Lusanovskoe oilfield.

Density (g/cm^3)	Effective viscosity at 20 °C (mPa×s)	Freezing point (°C)	Contents (% mass)		
			Oil	Resins	Asphaltenes
0.953	1014	−17	64.05	28.6	6.1

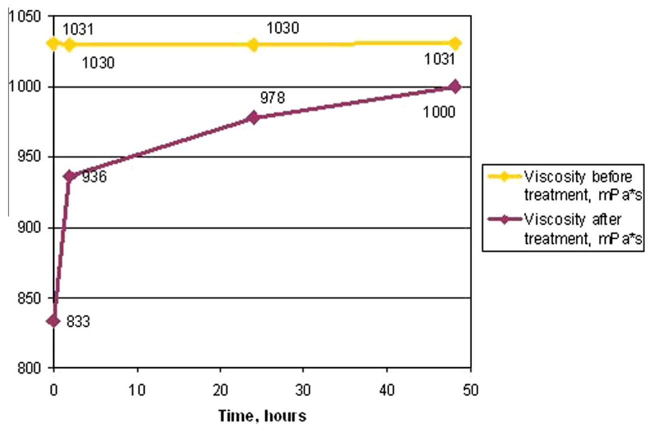


Fig. 2. Changes of the dynamic viscosity of oil within 48 h after ultrasonic treatment.

thermal effect of ultrasound the probe was kept in a water bath. The pressure was kept constant.

The oil used for the experiments was from the Lusanovskoe oilfield. Its properties are presented in Table 1.

We used a viscosity meter (SX-80) to measure the viscosity of the oil before and after treatment. According to the theoretical model described above the time required to destroy the intermolecular connections directly near the emitter is 30 s., however to ensure the destruction within the whole reactor at least 1 min is required. The treatment time was 3 min. A viscosity reduction was observed from 1 until 3 min of treatment. After ultrasonic processing the oil was monitored for a further 48 h. If the reduction of oil observed was because of destruction of the conglomerates, a rise of the viscosity should be observed after the recombination of these conglomerates.

The experiments revealed, that a pronounced deduction of viscosity is observed directly after ultrasonic treatment of viscous oil, however relaxation of this effect is observed within 48 h, after which the viscosity returns to its original values. The results of this experiment are presented on Fig. 2. The treatment time needed for the reduction of viscosity is in good agreement with the time calculated in the theoretical model above.

The estimations given above are in good agreement with field test results presented in Ref. [12], which proof that good results in terms of enhanced oil production were achieved in vertical wells when not less than one hour was spent when for the treatment. This is the time needed to destroy intermolecular connections in conglomerates in the oil and in the surface layer near the rock formation one meter away from the emitting surface of the downhole tool in order to clean the wellbore perforation zone.

This treatment time is economically justified for vertical wells, but is not suitable for horizontal wells because the length of the perforation zone in them may reach hundreds of meters.

Also it should be taken into account that the use of treatment results obtained from vertical wells is not always able to be extrapolated to possible results in horizontal wells, as there are a number of differences in the formation of the zones near the wellbore:

- The overall geological inhomogeneity affects the zone near the wellbore.
- The borehole is affected by stronger and more intensive deformation processes compared with a borehole of a vertical well.
- Technologies of drilling and finishing of vertical and horizontal wells differ markedly.
- One of the main peculiarities of perforation zones of horizontal wells is low pressure gradients.

Thus, for treatment of horizontal wells a sonochemical approach [11] should be used. This method enables us to decrease the treatment time due to the synergetic effect, which is achieved when a combination of acoustical and chemical treatment is used. In this case ultrasonic treatment not only contributes to the cleaning of the perforation zone and increasing the mobility of oil, but also to the penetration of the reagent into the formation and acceleration of the chemical reaction in the porous media of the formation.

3. Equipment

In the case of inhomogeneous layered reservoirs with low permeability, which are typical for Western Siberia, the decrease of oil production from horizontal wells can occur for the following reasons:

- The drilling fluid decreases the permeability of the horizontal area of the well by 15–25% after finishing.
- The formation becomes clogged during exploitation and so its effective porosity decreases, this results in a decrease in oil production of 10–35%.

Such wells are chemically treated nowadays, however the reagents often penetrate through regions of minimal resistance forming “tongues”.

Thus, in order to carry out effective treatment of horizontal wells specially designed equipment has been developed. Any technology of EOR for horizontal wells should be based upon the following guidelines:

- The formation intervals i.e. those which need treatment should be based on geophysical studies.
- In order to achieve synergetic effects the reagents need to be injected directly into the zone of acoustic treatment.
- The treatment should be selective in that only the problematic zones should be treated, this decreases the overall treatment time.

The ground and downhole equipment developed for sonochemical EOR has been significantly improved over the last years. The ground equipment includes an upgraded ultrasonic generator involving a unit for processing the information obtained regarding pressure and temperature in the borehole, this information is obtained from a downhole tool. The ultrasonic generator is matched to the ultrasonic downhole tool and easily adapts to changes in the technological load by controlling the voltage and the current which goes to the downhole tool. The ultrasonic generator can work in a pulse mode and can modulate the power. The operating frequency is 15–30 kHz, the output power is 10 kW. During the operation of the generator the following parameters can be monitored on the display: voltage, current, work/pause, frequency.

The downhole equipment includes a sonotrode, a system for injection of chemicals and a probe for acquiring geophysical data (temperature, pressure, flow). In order to use this equipment in horizontal wells the equipment complex must include a special

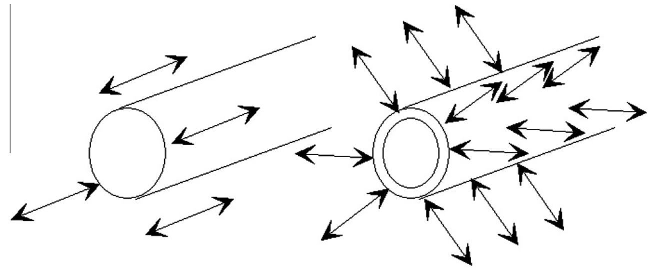
Fig. 3. Crossection of the cable with hydraulic channel and power and signal cores.

cable with a hydraulic channel for injection of technological fluids. The cable includes electrical cores to power the ultrasonic equipment (1.5 mm diameter) and the geophysical probe (4 signal cores to control the parameters of the process). The cable is an armored polymeric tube, the copper cores are nested within it. The cross section of the cable is represented in Fig. 3. The diameter of the hydraulic channel is 15 mm. The power up to 5 kW can be delivered through the cable. The cable can be also used for moving the ultrasonic downhole tool through the horizontal area of the well during the treatment. Apart from the injection of reagents directly to the zone of the acoustical treatment the cable can also be used to wash and clean the horizontal area of the well with technological fluid prior to and after the treatment.

The armor surrounding the cable protects it from external damage. It has the required breaking strength and torsional stiffness to be wound onto a drum on a geophysical truck. Photographs of the wireline truck constructed specially for sonochemical treatment of horizontal wells in the transport (a) and working (b) position are presented in Fig. 4.

During treatment of wells it is necessary to continuously process the data in order to choose the appropriate treatment modes and to adjust them during the operation. Complex geophysical downhole tools which are used during the treatment measure the following parameters:

- Pressure
- Temperature



(a) (b)

Fig. 5. Oscillation character in rod type (a) waveguide systems and push–pull system with a developed radiation surface (b).

- Natural radiation of the rock
- Flow of the fluid
- Magnetic location of the couplings
- Thermoconductive flow
- Resistance
- Soil/water content

The flow from the horizontal area can be measured before during and after treatment.

To select the optimal design of the sonotrode various waveguide systems were designed, modeled, manufactured and tested. Best results were achieved when push–pull type sonotrodes were used. The operation of such sonotrodes is based on conversion of longitudinal oscillations to radial, which are emitted from the sidewall. This leads to increase of the efficiency of the sonotrodes. The conversion is possible when the frequencies of radial and longitudinal oscillations are matched. In this case the character of the oscillation changes (Fig. 5). The operating frequency of the sonotrode used for sonochemical treatments of wells was 20 kHz.

All the above mentioned downhole equipment worked under the following conditions:

- Temperature: up to +150 C°
- Maximum pressure 60 MPa
- Acidic environment up to 12% with surfactants

Apart of construction and optimization of equipment a major factor which affects the result of sonochemical treatment is the chemical reagent employed. A composition called "IHN-Pro", was developed especially for this purpose in the Institute of Oil Chemistry of the Siberian Department of the Russian Academy of Sciences. It contributes significantly to the growth of oil production

(a) (b)

Fig. 4. Photographs of the wireline truck for sonochemical treatment of horizontal wells in transport (a) and working (b) position.

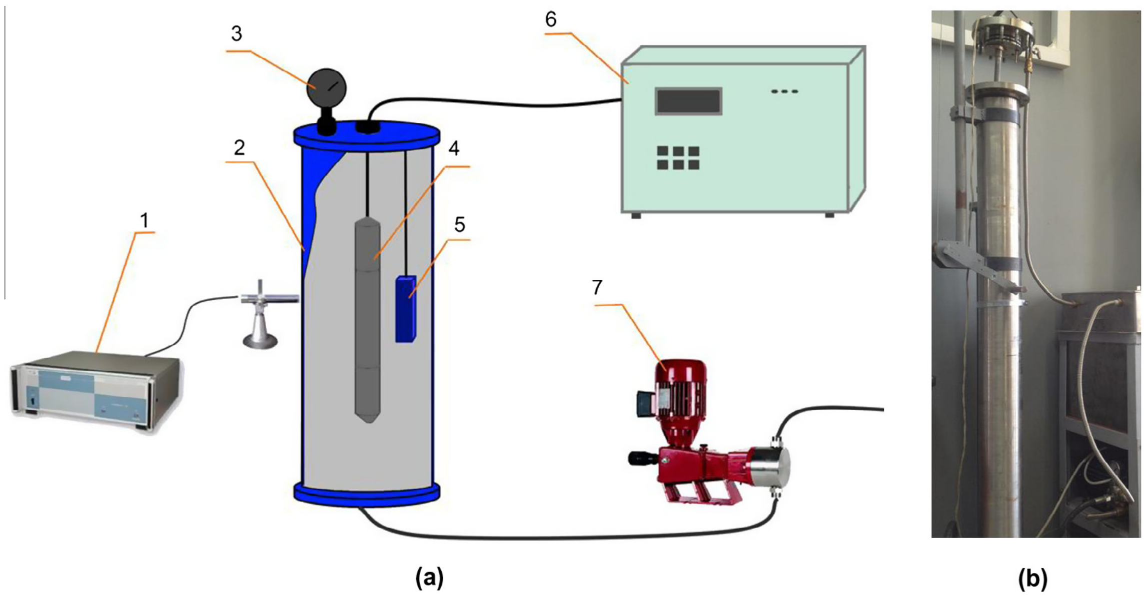


Fig. 6. Scheme of the laboratory equipment for study of the sonochemical effect on the core sample (a) and its photograph (b).

from a range of underground formations with different geological and physical characteristics.

The reagent is compatible with mineralized waters of the reservoir, has a low freezing point and is not explosive. It is based on a combination of surfactants, an alkaline buffer system and polyhydric alcohols and its properties are presented below.

- Density at 20 °C – 1.21 kg/m^3
- Viscosity at 20 °C – 19 mPa×s
- Working temperature range 10–250 °C
- pH 4.5–6.5

The effect of the reagent on the formation during ultrasonic treatment has been studied under laboratory conditions as described below. To test the efficiency of sonochemical treatment core samples of a sandstone reservoir were used. The permeability of this core samples was 40 mD. The samples were saturated with oil in the device UN-3 and placed in the laboratory equipment shown in Fig. 6. The equipment consists of a device for amplitude measurement on the sidewall of a barochamber (1), a chamber with high pressure (2), a manometer (3), a sonotrode with magnetostrictive transducers (4), a core sample holder (5), an ultrasonic generator (6) and a pump (7).

In order to estimate the effect of the ultrasonic treatment on core sample cleaning using IHN-Pro the chamber was filled with

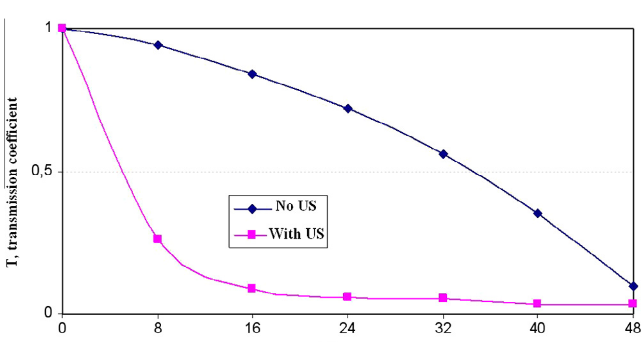


Fig. 7. Dependence of the transmission coefficient on the time of reaction in a barochamber with a core sample saturated with oil with and without ultrasonic treatment.

the reagent. The concentration of the oil in the reagent was monitored with time both with and without ultrasonic treatment. The concentration of oil in the reagent was estimated based in the changes of light transmission coefficient. The measurements were carried out using an SF-2000 spectrometer. The spectrometer measures the ratio of two luminous fluxes: the luminous flux, which passed through the sample and the control luminous flux. The transmission coefficient is then calculated using Eq. (11):

$$T = \frac{I - I_t}{I_k - I_t} \tag{11}$$

where I is proportional to the luminous flux through the sample, I_k is proportional to the control luminous flux, I_t is proportional to the "dark" luminous flux. The control luminous flux was the luminous flux through IHN-Pro in its original state. Fig. 7 shows the dependence of the transmission coefficient on time with (sonochemical) and without (chemical) ultrasonic treatment.

As shown in Fig. 7 acoustic treatment increases the activity of IHN-Pro and, consequently, leads to a decrease in the time needed to treat the wellbore perforation zone.

4. Field test results

In order to develop the optimal methodology of field test operations a number of tests have been carried out. First of all, the efficiency of the method was compared with the efficiency of ultrasonic treatment alone i.e. using the same methodology but without IHN-Pro. The sequence of operations employed has been described elsewhere [12] and the equipment referred to above was used. The time of ultrasonic treatment alone for 1 m of formation was 1 h as above. For sonochemical treatment the time of ultrasonic exposure was decreased to 30 min.

The comparison has been carried out on vertical wells in Western Siberia (WS) and the Samara Region (SR). More than 100 ultrasonic and sonochemical operations were performed in the period from 2010 to 2013. Table 2 shows average oil production before and after ultrasonic and sonochemical treatment, and illustrates the changes in the production of the well in 3 month after treatment.

On average after sonochemical treatment the oil production was higher and also the duration of the effect was longer. This

Table 2
Effect of ultrasonic and sonochemical treatment on the oil production.

Region	Type of treatment	Time of ultrasonic treatment (min)	Oil production (tons/day)		
			Before treatment	After treatment	3 month after treatment
Western Siberia	Sonochemical	30	3.92	9.1	8.4
	Ultrasonic	60	3.92	8.32	7.7
Samara region	Sonochemical	30	8.4	19.8	15.8
	Ultrasonic	60	8.4	18.6	11.5

can be due to the fact that materials that clog the well (colmatants) are removed during sonochemical treatment from all the pores, including the smallest. Under the influence of ultrasound the chemicals penetrate into the smallest pores due to the sonocapillary effect. The combined treatment gave the better effect even though the treatment time in the sonochemical method was only half of that using ultrasound alone.

Normally ultrasonic and sonochemical treatment of the well bottom is done during "down-time" in an oil well operation i.e. during workover of the well. Conventional down-time is often accompanied by optimization of the pumping equipment. In order to differentiate between the effects of ultrasound and normal workover we have measured the influence of ultrasonic treatment and workover on the changes in the productivity factor of the oil well and water cut i.e. the percentage of water in the recovered well fluid. Ultrasonic treatment leads to an increase of the productivity factor by 39% and decrease of the water cut of the well by 5% on average. Whereas in wells where only the optimization of pumping equipment was carried out there was a drop in the productivity factor of 5.6% and an increase in the water cut of 1.5%. The tests indicated that the success rate of the ultrasonic treatment of vertical wells reaches 90% and the increase in oil production is in the range of 40–100%.

We have only begun work on the sonochemical treatment of horizontal wells and so we do not have sufficient information to provide a full statistical analysis. However, the results obtained to date allow us to take an optimistic view about the potential of this technology. Thus far we have treated 3 horizontal wells in sandstone reservoirs in Western Siberia. The time of ultrasonic treatment of 1 m of the formation after injection of the reagent was 15 min. Before and after sonochemical treatment of the well geophysical studies of the well were carried out. Based on the information received the zones for sonochemical treatment were determined. The treated area was 200–300 m long, the productive formation had a porosity of 0.27, the permeability was 0.515 μm^2 and oil saturation was 0.67.

As a result of sonochemical treatment the production of fluid and production of oil from all three treated wells grew. On average the production of fluid increased from 51 to 72 tons per day, and the production of oil from 23 to 33 tons per day. In comparison with the sonochemical treatment of vertical wells in the same region the treatment of horizontal wells definitely improved oil production but not to the same extent as with vertical wells and the change of the water cut after treatment was negligible.

From such a promising beginning it is clear that further research is required in order to optimize the equipment and the methodology of sonochemical treatment of horizontal oil wells. Our first field tests show a marked improvement over currently favored hydraulic methods of secondary intensification that can filter only 10–15% of the formation due to irregularity of permeability along the well. The reagents used for treatment of horizontal wells are acids, oxidants, enzymes and chelates. Potentially all of these reagents and many more may be used for sonochemical treatment.

5. Conclusions

- Experimental results and theoretical estimations show that the optimal treatment time of ultrasonic enhanced oil recovery EOR in vertical wells is 60 min. For horizontal wells this time must be shorter.
- Laboratory experiments have shown that ultrasound can enhance the effect of chemicals used to improve the performance of vertical wells and to treat the wellbore perforation zone of horizontal wells.
- Field tests of the sonochemical method of EOR in vertical wells have shown a significant improvement in oil production, in a number of cases the production coefficient increased and the water cut decreased after sonochemical treatment.
- We have developed equipment for the sonochemical treatment of horizontal wells, which consists of a specialized wireline truck, an ultrasonic generator, a cable with a channel for injection of chemicals, geophysical equipment for controlling the process and a downhole ultrasonic tool with a "push–pull" waveguide system.

Acknowledgments

A part of this research was carried out as part of the activities within a project funded by the Russian Ministry of Education and Science, Contract No. 14.527.12.0002.

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