SPE-192564-MS

The First Comprehensive Study of Tracer-Based Technologies in Reservoir Conditions

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This paper was prepared for presentation at the SPE Annual Caspian Technical Conference and Exhibition held in Astana, Kazakhstan, 31st October – 2nd November 2018.

Abstract

At the present time, it has become progressively more common for countries around the world to use tracer's methods of production logging in horizontal wells. The utilization of tracers/markers doesn't require well intervention with Coiled Tubing or tractors in order to obtain qualitative and quantitative data for inflow of reservoir fluid per interval. Tracer/marker technologies is a quantum leap in production logging as it is potentially capable of producing ten (10) to twenty (20) times more downhole data in a period of several years. The knowledge of downhole production in dynamics of years helps in developing oil and gas fields, locating neighboring wells to be drilled, optimization of horizontal lateral length or even a number of fracturing stages per well.

The principal difference between these technologies and traditional methods of well surveys (WS) is their ability to monitor the operation of ports or intervals in a well over a long period of time, with a significant reduction in resources involved, a reduction in costs and an enhanced production safety. However, the question remains whether the claimed specifications of tracer technologies correspond to the actual ones. Sometimes, the decision on the implementation of tracer PLT is made with weak quality assurance measures causing subsequent compromise of project results as a whole.

The value of this article is the development of a methodology for a comprehensive assessment of the operability, reliability and accuracy of operation for various tracer technologies available in the market. Based on results of this research being conducted, the article suggests recommendations from producer companies that can be used as technical criteria when selecting a contractor for tracer studies.

Introduction

In retrospective, there has been a noticeable interest in tracer research in the world, however very few materials have been published on tests that confirm or deny the claimed advantages of these technologies.

Often, oil and gas companies make decisions to use tracer technologies without any testing and based only on the reputation of the supplier company as well as the duration of its presence in the market or value.

The reason for this may be the lack of unified test methods, as well as the experience of sharing best practices between oil producing companies. Tracer technologies are a relatively new subject in the field of well research, therefore, it is necessary that the approach in the assessment of technologies will be on the basis of objective results.

Classification of tracer technologies for the study of wells

Tracer (marker) methods of research use the following marking technologies:

- Chemical water and oil-soluble reagents (fluorescent, ionic and organic tracers);
- Quantum markers-reporters;
- Chemical DNA tracers.

Fluorescent and ionic tracers as indicators for the study of oil-and-gas bearing reservoir have been widely used since the last century. Despite the fact that these reagents are of relatively low cost, their drawback is the difficulty in reliable quantification of well flow log during monitoring, as well as the limiting number of tracer signatures/codes to 5-7.

Quantum markers-reporters are polymeric monodisperse microspheres. Their identification is being carried out by the method of flow cytometry and using algorithms of machine learning [1, 2, and 3].

Chemical DNA tracers are long polymeric molecules consisting of repeating blocks – nucleotides. Identification of these tracers is conducted by using liquid chromatography in combination with mass spectrometry [4].

A general comparison of tracer technologies is presented in Table 1.

Table 1—Comparison of tracer technologies for the study of wells

№	Parameter	Chemical water and oil-soluble reagents			Chemical DNA tracers	Quantum markers-reporters
		Fluorescent tracers	Ionic tracers	Organic tracers		
1.	State	Dry powder/liquid			Polymer	Polymer matrix
2.	Type	5-7 (sodium fluorescein, disodium salt of eosin, erythrosine, rhodamine)	5-7 (ammonium rhodanate, sodium, carbamide, urea, sodium nitrate, ammonium; thiourea)	3-4 alcohols (isopropanol, butanol), isomers fluorobenzoic acid	No data	60 and above
3.	Water/oil solubility	Very soluble in water	Very soluble in water	Soluble in both oil and water	Soluble in both oil and water	Soluble in both oil and water
4.	Resistance in reservoir conditions	Mean	Mean	High	High	High
5.	Marker identification method	Fluorescence microscopy	Photometry, electron-paramagnetic resonance spectroscopy	Chromatography	Liquid chromatography, mass spectrometry	Flow cytometry
6.	Analyzer	Fluorate, fluorescence microscope	Fluorate, photoelectric colorimeter	Chromatograph	Chromatograph, mass spectrograph	Analytical hardware and software complex GEOSPLIT
7.	Sensitivity	From 1 mg/t	No data	From 1 mg/t	No data	No restrictions
8.	Additional restrictions	1. Absence of mono-dispersion. 2. "Slushed" by polar organics. 3. Presence of similar substances in the reservoir	Difficulties in quantitative determination due to low accuracy.	1. High duration of research. 2. Difficulties in application for industrial scales.	-	-

In addition to the type of tracers used, it is possible to classify tracer technologies by the following criteria:

1. By the type of wells:
   - Vertical;
   - Horizontal;
   - Slanted;
   - Multi-lateral.
2. By the method of introducing tracers into the well (reservoir):
   - For new wells during well completion process (in downhole casings, filters of special constructions, cassettes);
   - For old wells using cassettes being deployed downhole with coiled tubing or work-over rig and anchored along wellbore;
   - Application of a marked polymer-coated proppant under a multifrac;
   - Injection of tracers with liquid (HFT-fluid, acid solutions).
3. By the method of lab analysis:
   - Manual (for example, using microscopes);
   - Automated (with the use of modern software and hardware systems and also self-teaching software).

Features of the application of quantum markers-reporters

The technology is based on the use of markers-reporters from quantum dots embedded into the polymer coating of a proppant being utilized to fasten cracks during a multi-stage hydrofrac (Figure 1) or their use in packaging arrangements of completion (Figure 2).

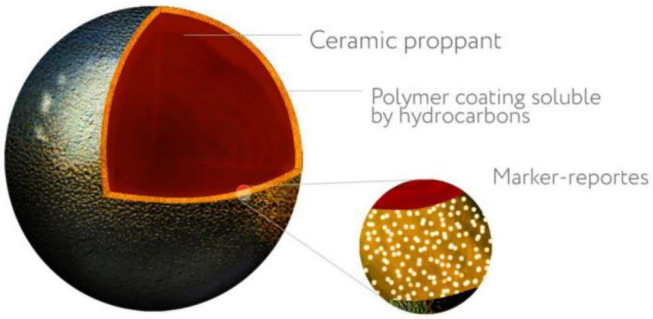

Figure 1—Marked polymer-coated proppant

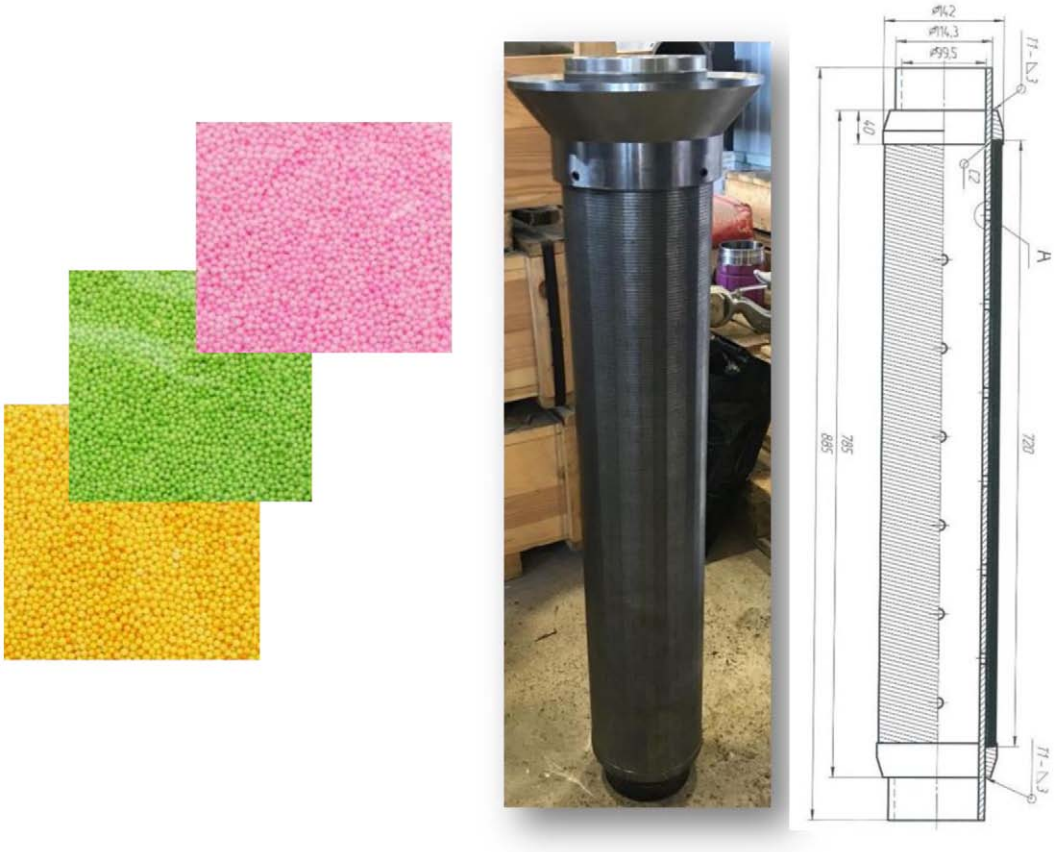

Figure 2—Cassette filled by polymer granules with markers to be deployed in a horizontal section

In the process of operation, the markers-reporters are washed out by the flow of reservoir fluid for a long period of time. When collecting samples from the wellhead and subsequent laboratory tests, the analytical hardware-software complex GEOSPLIT determines the concentration of markers for each code (Figure 3), that allowed us to estimate the quantitative distribution of oil and water phases for each horizontal section.

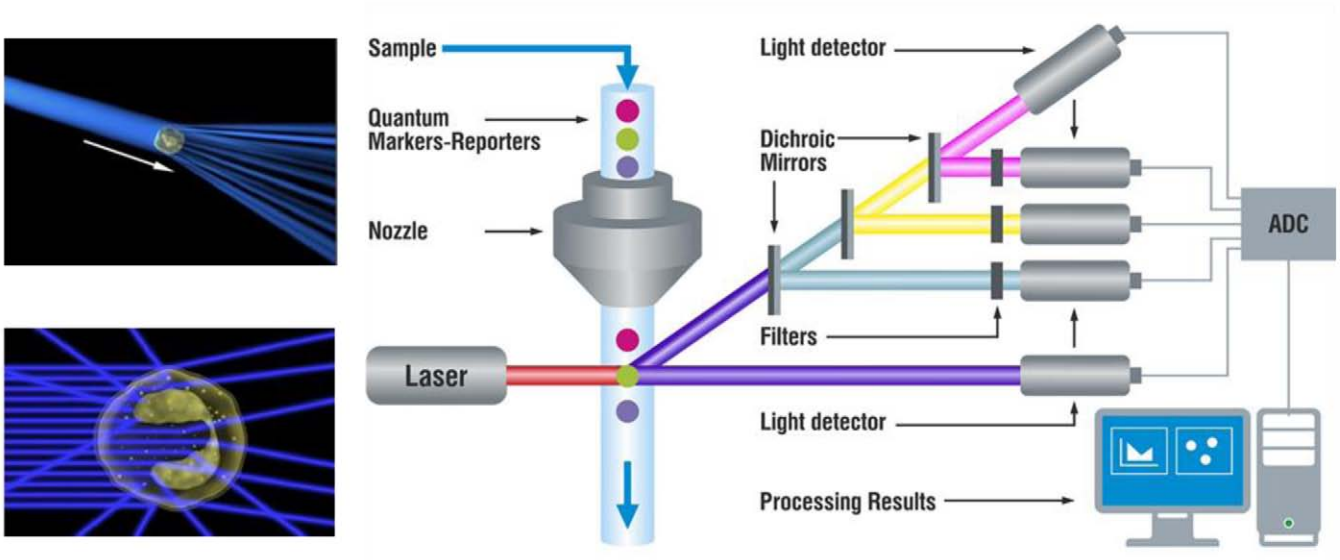

Figure 3—Analytical hardware-software complex GEOSPLIT, implementing the identification of markers-reporters by a flow cytometry method

The main advantages of using quantum markers-reporters include:

1. Monodispersity of markers by size.
   The lack of monodispersity of tracers introduces considerable margin of error for reliable quantitative analysis, since particles of different sizes have different sedimentation rates and, as a consequence, different relative flow velocities in the wellbore. Particles of small sizes will be removed by the fluid flow faster compared with larger particles. In addition, particles of different sizes differ in their ability to move with the reservoir fluid in the reservoir.
2. Automated identification of markers in samples of reservoir fluid.
   The identification of markers is carried out with automated software and hardware complex in the mode of piece-by-piece analysis without the use of microscopes. When analyzing samples, a strict number of markers-reporters is identified in as pieces per each sample which ensures high accuracy of the studies and eliminates errors related to human factor.
3. Uniform output of markers for an extended period of time.
   Markers-reporters, sewed into the polymer matrix of the proppant or granulate, ensure the stability of the concentration release from the polymer coating.
4. A large number of signatures (codes) of markers.
   At present, it is possible to synthesize more than 60 unique signatures of markers for hydrophilic and hydrophobic polymer coatings that allows to perform the diagnosis and monitoring of 30 horizontal sections simultaneously.
5. No restrictions for the use of markers in reservoir conditions.
   Markers-reporters show high physical-and-chemical stability, as well as resistance to the influence of aggressive media and reservoir thermobaric conditions.

Internal Testing

Work experience with different customers in different regions has shown that additional tests are needed in order to confirm the operation of the technology including the claimed advantages.

To this end, GeoSplit LLC conducted a wide range of laboratory tests of various marker systems, including:

1. basic tests for the physical-and-chemical stability of markers and the polymer matrix (temperature stability, stability of the concentration of markers' separation, acid resistance, resistance to hydrogen sulfide);
2. tests for determining the basic permeability (conductivity) of a proppant pack;
3. crash test of study on the effect of proppant destruction on the intensity of markers' separation;
4. marked proppant compatibility tests for HFT-gel;
5. tests for the transition of markers via interphase water-hydrocarbon boundary.

Basic tests on the physical-and-chemical stability of markers and the polymer matrix

Tests on temperature stability and stability of the concentration of markers' separation in time. The tests were carried out in a column filled up with proppant through which the target fluid (water, oil) was passed at various temperatures and with a fixed flow rate (Figure 4), after which fluid samples were collected and analyzed in the laboratory.

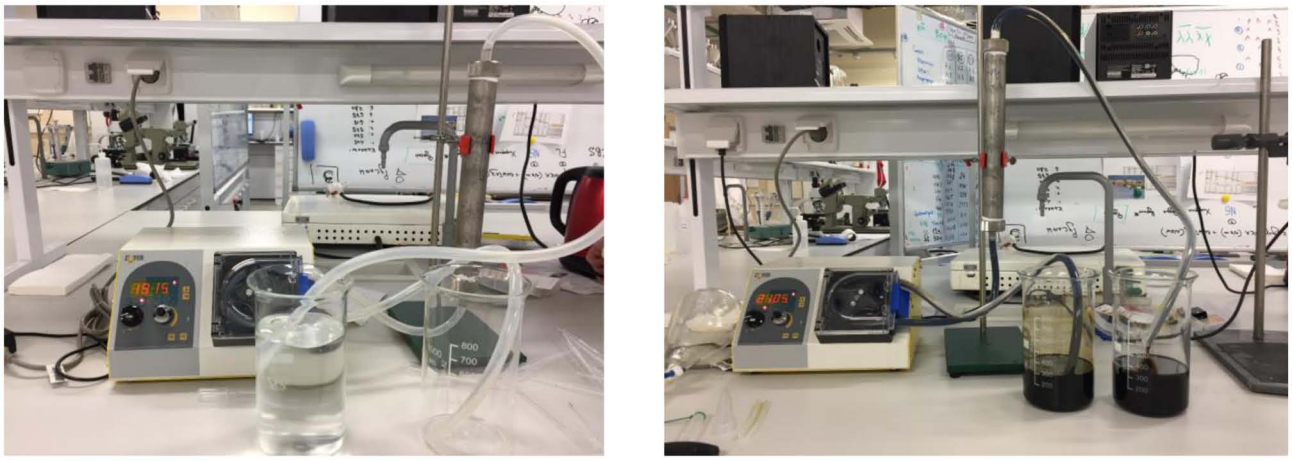

Figure 4—Laboratory facilities for testing with water (left) and oil (right)

Figures 5-6 show the tests results on the stability of the concentration of markers' separation in time at different temperatures and with a fixed flow rate for the hydrophilic (HP) and oleophilic (OP) marked proppant.

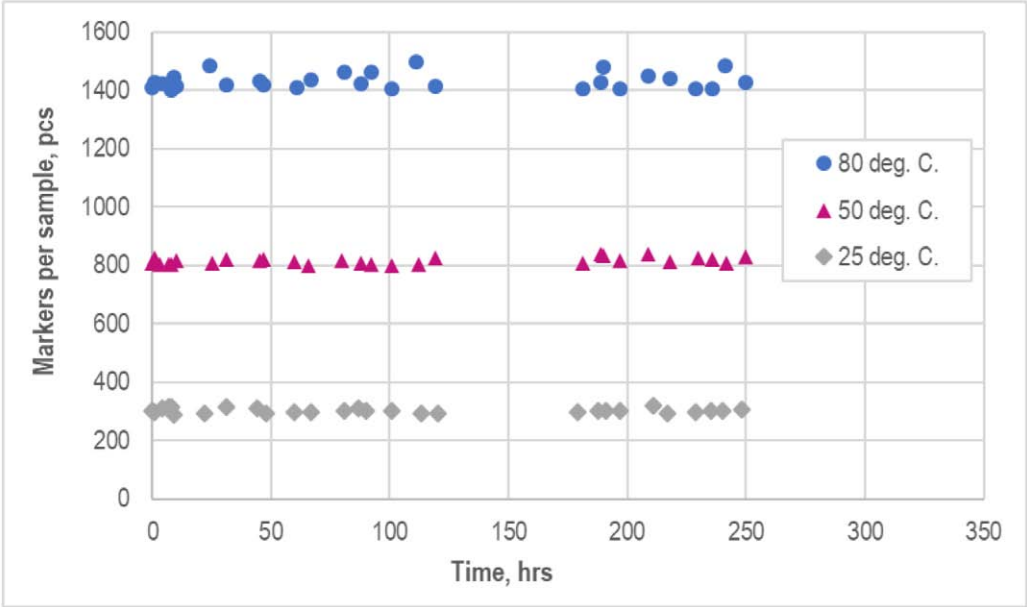

Figure 5—Dependence of the number of markers released from the hydrophilic proppant (HF), on the time at different temperatures and a fixed flow rate

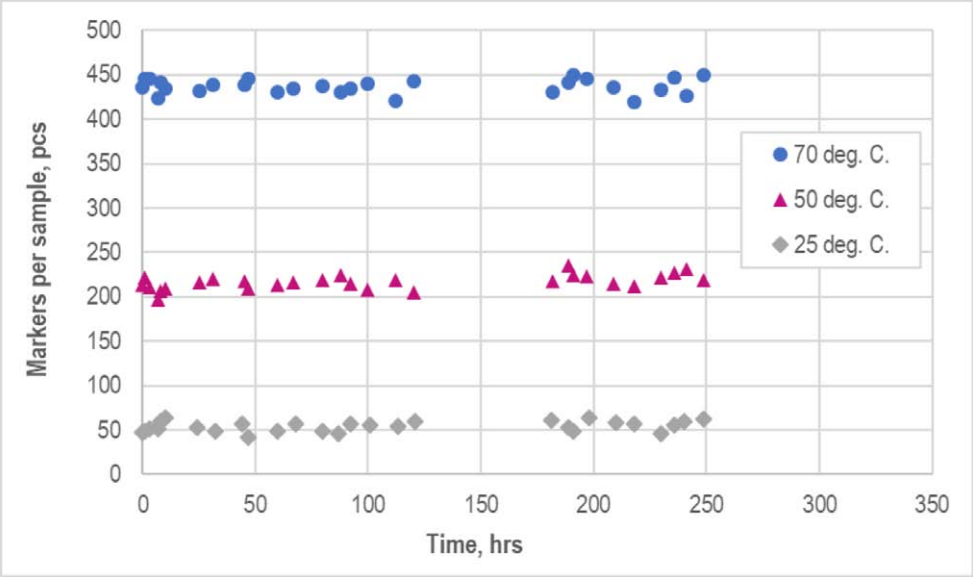

Figure 6—Dependence of the number of markers separated from the oleophilic proppant (OF), on the time at different temperatures and a fixed flow rate

Based from tests results, there has been a stable concentration of markers' separation from the polymer coating of the proppant at different temperatures.

Tests for researching the dependence of markers' separation in water-oil medium at different values of water encroachment. The tests were carried out in a similar manner in a column filled out with proppant through which the water-oil mixture was passed at different values of water encroachment, after which, fluid samples were collected and analyzed in the laboratory.

Figures 7-8 reflect the results of a study on the dependencies of markers' separation in water/oil phase on the fluid flow rate at different values of water encroachment.

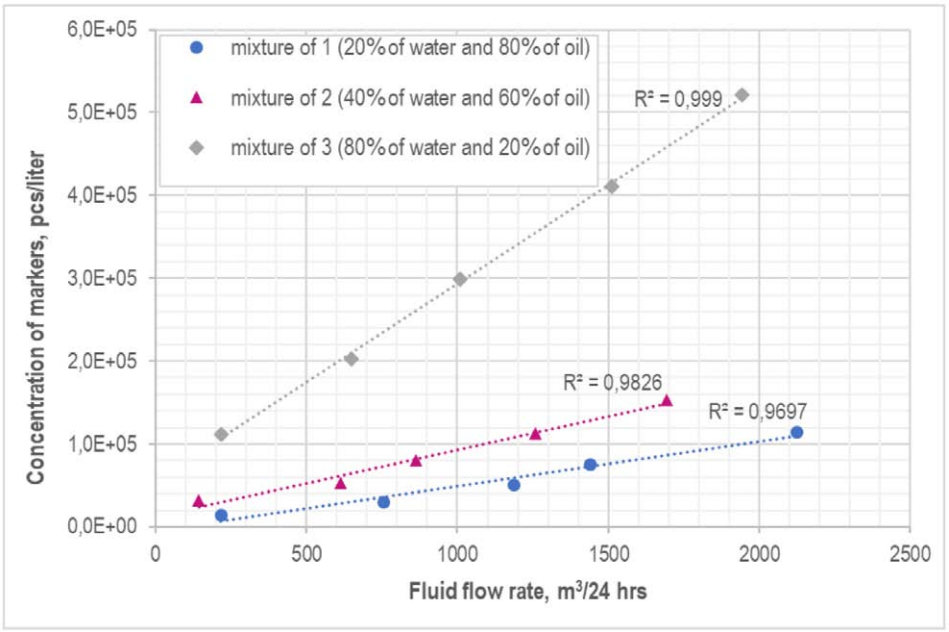

Figure 7—Dependence of the number of markers separated from a mixture of proppants in the water phase, on the fluid flow rate at different values of water encroachment: mixture of 1 − 20% of water and 80% of oil; mixture of 2 − 40% of water and 60% of oil; mixture of 3 − 80% of water and 20% of oil

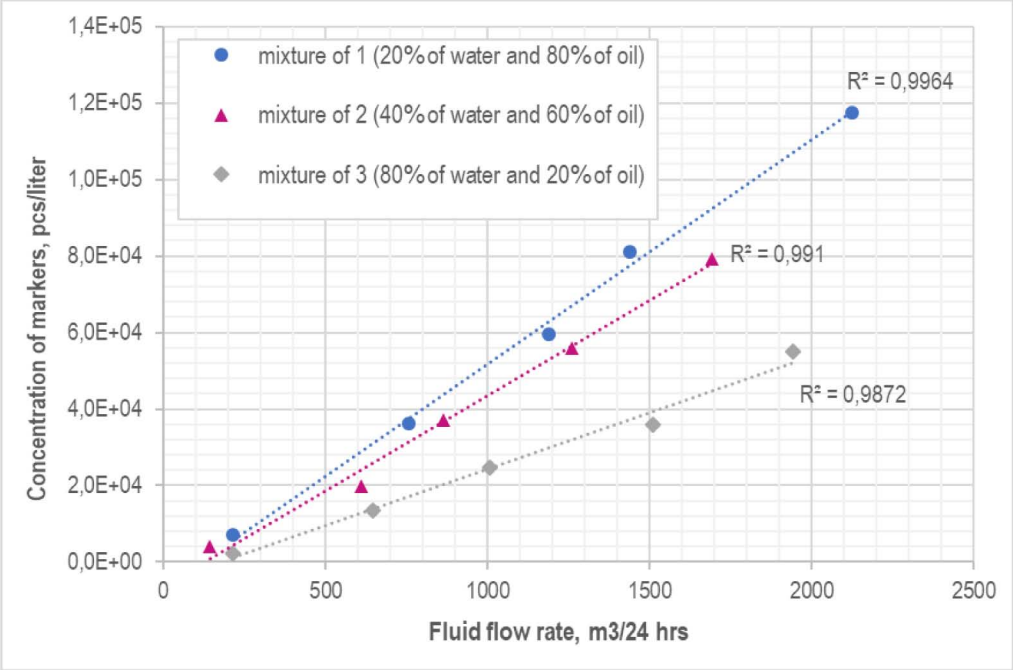

Figure 8—Dependence of the number of markers separated from the proppant mixture in the oil phase, on the fluid flow rate at different values of water encroachment: mixture of 1 − 20% of water and 80% of oil; mixture of 2 − 40% of water and 60% of oil; mixture of 3 − 80% of water and 20% of oil

The experimental results have proven that the character of the separation of markers from the polymer coating, depending on the fluid flow rate, including the water-oil mixture at different values of water encroachment, is linear, which makes it possible to unambiguously quantify the distribution of horizontal wellbore inflow profiles.

Tests on acid resistance. The tests were carried out by gravimetric determination of the change in sample mass of hydrophilic (HF) and oleophilic (OF) proppants after treatment with working acid solutions:

- mixture of concentrated hydrochloric and hydrofluoric acids;
- hydrochloric acid solution.

The results of acid resistance tests are shown in Table 2.

Table 2—Test results on acid resistance

№	Type of acid	Relative change in mass of the marked proppant after treatment,%	The requirement of the quality standard (GOST R 51761)	Compliance with the quality standard
1	A solution of a mixture of hydrochloric and hydrofluoric acids with a mass ratio of 4:1	7,0	8,0	Yes
2	Hydrochloric acid with a concentration of 15%	0,9	1,0	Yes

According to test results, it was found that the marked proppant shows a low solubility in acids and meet the requirements of quality standards.

Tests on hydrogen sulfide resistance. The tests were carried out by saturating water and oil with hydrogen sulphide by bubbling through them the gas obtained from the reaction of iron sulphide (II) with hydrochloric

acid and passing the target fluid saturated with hydrogen sulfide through a proppant column in order to determine the mass loss of the proppant.

The results of testing for resistance to hydrogen sulphide have shown that the weight loss is:

- 0.07%, in a solution of H_2S-saturated water;
- 0.04%, in a solution of H_2S-saturated oil.

A reduced loss of mass indicates the inertness of the polymer coating in relation to hydrogen sulphide.

Figures 9-10 show the dependence of the number of markers separated from the proppant on the time of fluid passage at a fixed flow rate of 200 m^3/day.

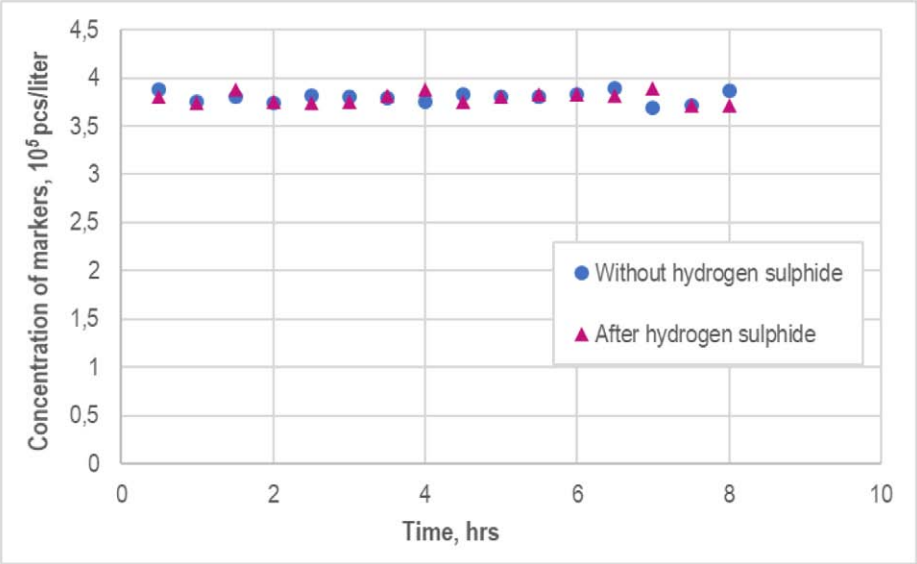

Figure 9—Dependence of the concentration of markers in a water sample on the time of passage through a marked proppant of H_2S-saturated fluid

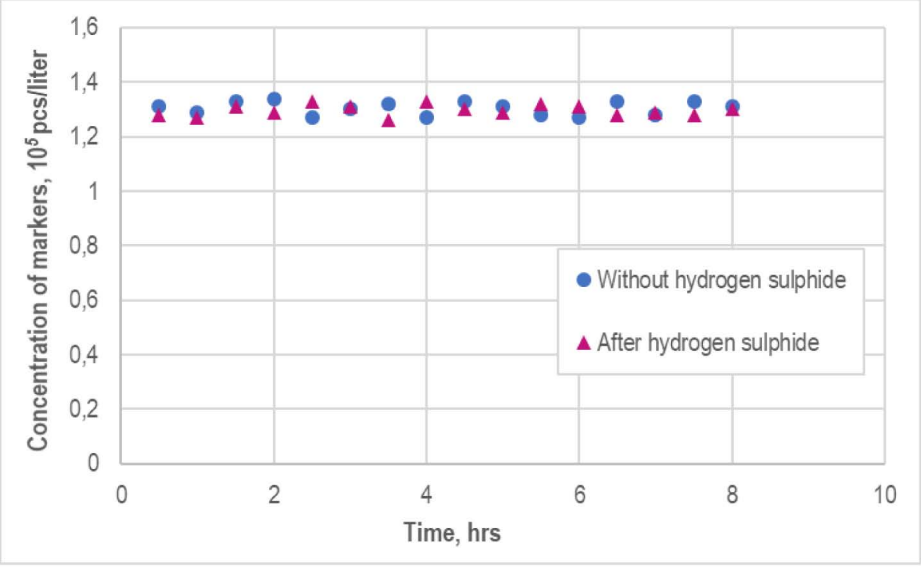

Figure 10—Dependence of the concentration of markers in the oil sample on the time of passage through the marked proppant of H_2S-saturated fluid

The experiments have shown that the polymer coating of the marked proppant is chemically resistant to hydrogen sulfide. Also, the presence of hydrogen sulphide in the reservoir fluid does not affect the separation of markers from the polymer coating of the proppant.

Tests on the determination of the basic permeability (conductivity) of a proppant pack

This experiment is based on the technique for measuring the long-term specific conductivity according to ISO 13503-5. The tests were carried out with marked proppant produced by the company GEOSPLIT (fraction 30/50 GS) and with the conventional uncoated proppant (fraction 30/50 UP) in order to compare the permeability and conductivity of both proppants in the following conditions:

- temperature of 100 °C;
- geostatic pressure of 300, 450, 550 and 700 atm.

The dependences of the permeability and conductivity of proppant packs on the geostatic pressure are shown in Figures 11-12.

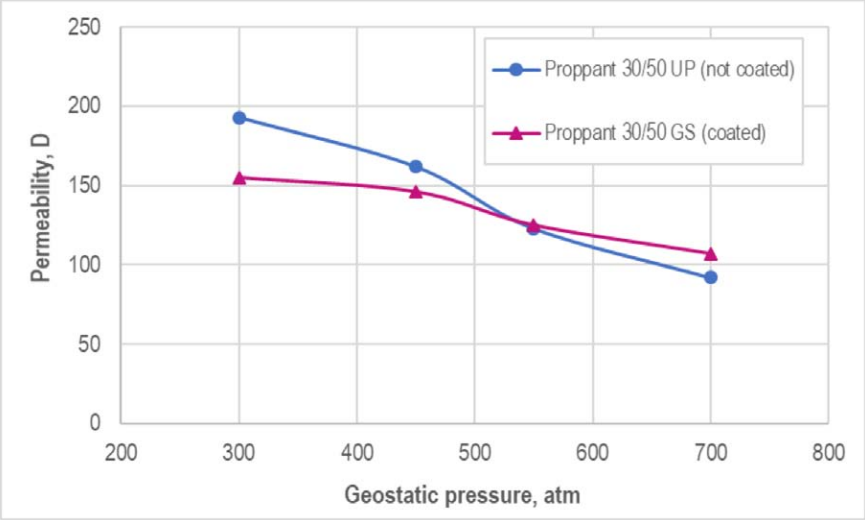

Figure 11—Dependence of permeability of proppant packs on the geostatic pressure

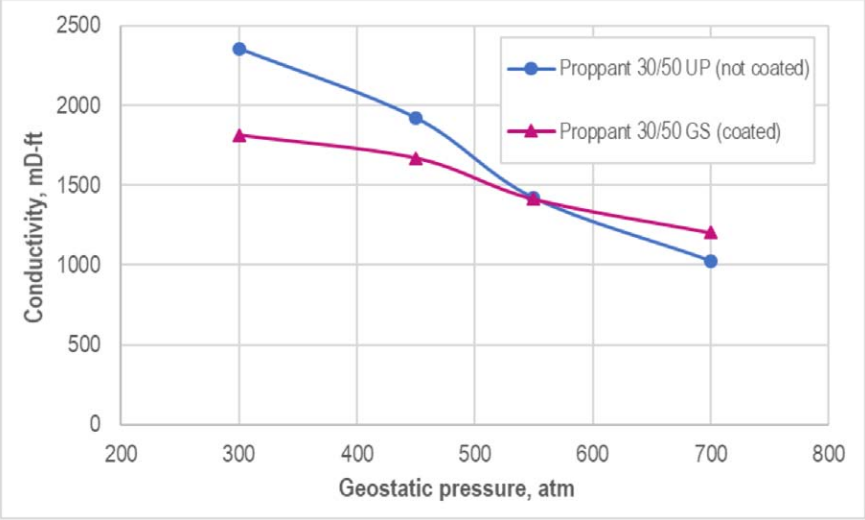

Figure 12—Dependence of the conductivity of proppant packs on the geostatic pressure

The results show that with geostatic pressures of less than 550 atm, the permeability and conductivity of the proppant pack for the marked proppant of 30/50GS is lower than that of the uncoated 30/50 UP proppant that is a typical property of polymer-coated proppants.

However, at geostatic pressure of 550 atm and above, the conductivity of the proppant pack of the marked proppant became higher. This is possibly due to the fact that in the polymer-coated proppant, the contact area increases as compared with the conventional one. The presence of a polymer film in the proppant at high pressure and temperature leads to the adhesion of individual grains to each other, therefore, its destruction occurs in a smaller amount. Hence, the fragments of the uncoated proppant are more likely to plug the inter-porous space and reduce conductivity.

It should also be noted that the permeability reduction of the polymer-coated marked proppant reached 31%, and of the uncoated proppant 52%, which is an additional advantage for using polymer-coated proppants.

Crash test on study of the effect of proppant destruction to the intensity of separation of markers

The crash test was carried out by means of a marked proppant grinding with a mechanical agitator at a rotation speed of 2400 rpm. Samples of mixtures of the destroyed and undestroyed proppants were then prepared; the mass fractions of the destroyed proppant were 0, 5, 10, 15 and 20%. Each sample was filled with distilled water with the same mass as the weight of the sample.

The results of the experiment are shown in Figure 13.

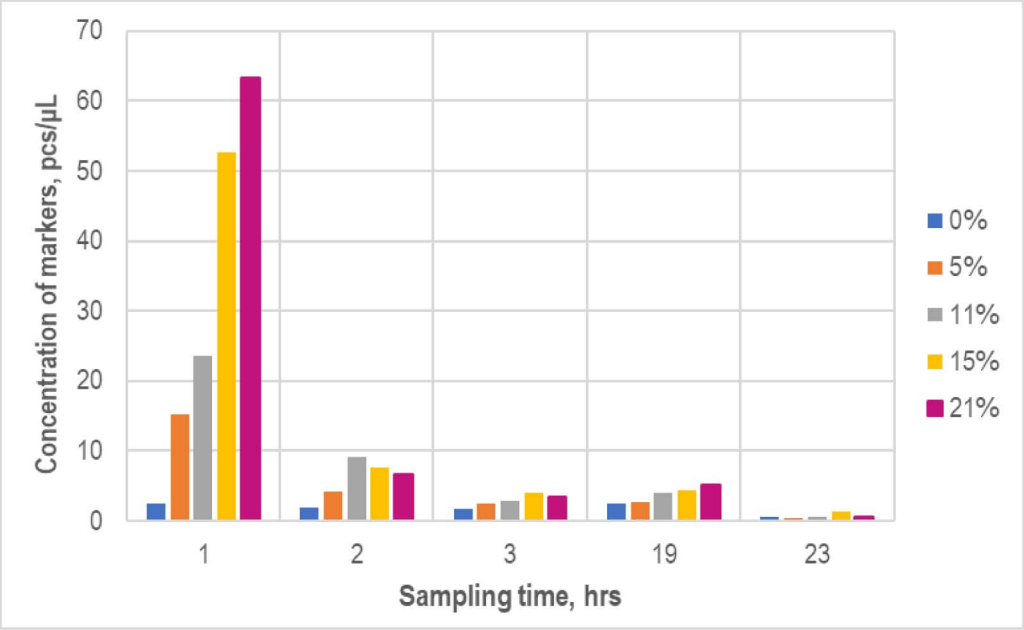

Figure 13—Dependence of concentration of markers on the time at various mass contents of the destroyed proppant

The experiment had indicated that the physical destruction of the proppant with markers leads to a rapid washout of markers from the destroyed grains, as a result of which there is a short-term jump in the content of markers in samples. At the same time, the described effect quickly disappeared due to the removal of a significant part of markers by the fluid. Thus, partial destruction of the proppant in reservoir conditions can be registered in one of the single samples, but does not significantly affect the content of markers in long-term studies.

Tests on the compatibility of the marked proppant with HFT-gel

The purpose of the tests was to determine the effect of the polymer coating of the marked proppant on the stability and profile of the fracturing HFT-gel using oxidative destructors at a temperature of 110 °C.

During these tests, the proppant was added directly to the linear gel and mixed for 5 minutes. Then, the pH-parameter was measured and afterwards the gel was cross-linked with proppant, and the pH-parameter was measured again. Further, the cross-linked gel was filtered out of this mixture in order to determine its rheological properties.

The results of the tests are shown in Figure 14.

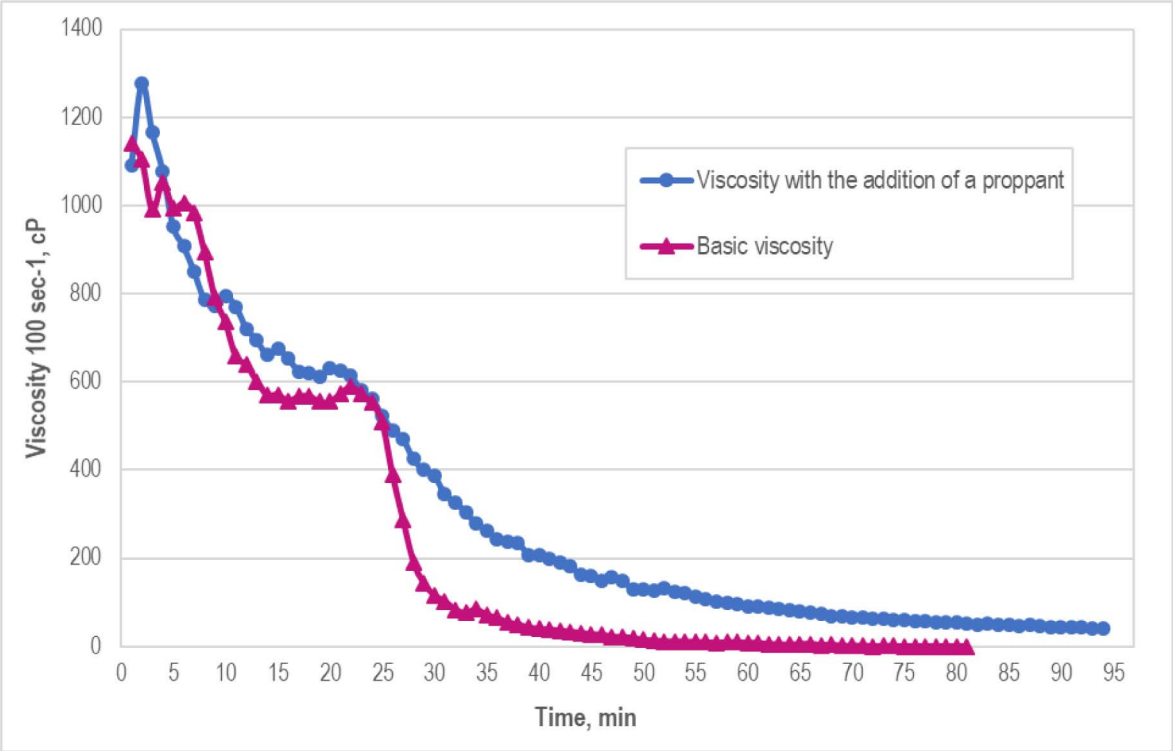

Figure 14—Test for compatibility of the proppant with markers and water-based HFT-gel with the addition of a capsized destructor

According to the obtained results, the addition of a marked proppant to a water-based HFT-fluid does not lead to its physical, chemical and rheological changes, but it slightly reduces the activity of the oxidative destructor, a consideration which should be taken into account when designing a HFT-design.

Tests for the transition of markers through the interphase water-hydrocarbon boundary

One of our Customers raised the question about possible transfers of markers from hydrocarbon phase to water phase and vice versa. As a specific hydrocarbon, hexane was used as one of the main constituents of oil and is an oil model for all properties (polarity, density, etc.).

The graphic scheme of the experiment is presented in Figure 15.

Markers were introduced in hexane, after which the sample was divided into two (2) parts. The first sample was not subjected to any action. The second sample was transferred to a centrifuge tube, filled with water of the same volume and scrolled in a centrifuge at 3000 rpm for 25 minutes. After centrifugation, hexane (upper phase) was collected, and water was processed in an ultrasonic bath (to lift the markers from the tube's bottom). The number of markers in hexane from the first sample and in the water from the second sample was measured using the Analytical hardware-software complex.

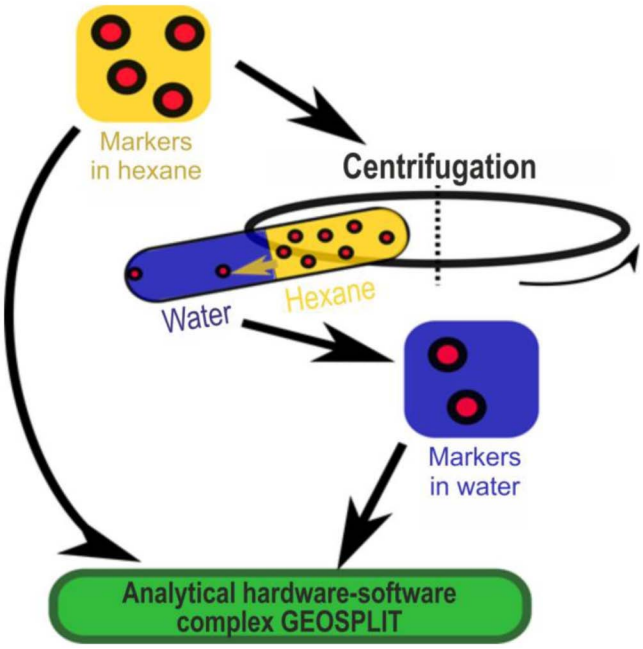

Figure 15—Graphic scheme of the experiment

During these experiments, significant non-equilibrium conditions were applied on the water-hexane system (centrifugation at acceleration of at least 1000 g for 25 minutes), but as a result, only a negligible share (0.04) of markers passed into the water phase. This allows us to draw a conclusion on the practical absence of the transition of markers from the hydrocarbon to the water and vice versa.

External Independent Tests

The key parameter for conducting external tests with the participation of customers and independent experts is the verification of the technology for accuracy of the quantitative determination of markers.

Tests on accuracy of the quantification of markers were conducted with the involvement of two customer companies (for the proppant solution and for the polymeric granules with markers) independently with each other. The tests were carried out according to the following procedure:

1. The manufacturer of markers-reporters (hereinafter referred to as the contractor) has provided for the independent commission of the customer samples of proppant with markers / polymeric granules with markers of 5 different signatures (codes).
2. The Commission, in the absence of representatives of company manufacturing marker-reporters, has prepared several mixtures containing different combinations of signatures, at the same time, samples of each signature were weighed in a laboratory scale; the measured mass was known only to commission members. Each sample of the mixture was signed with a conventional cipher.
3. Samples of mixtures were transferred to the research laboratory of markers manufacturer. In the presence of the commission, water containers (2% KCl solution) of 1 liter each were prepared, after which the mixture samples were placed in appropriate containers.
4. Next, each container was mixed by a mechanical agitator, after which a laboratory analysis of liquid samples taken from the containers was carried out.

The results of the comparison of data determined by the contractor and the commission's actual data for the marked proppant are presented in Table 3, for the polymer granulate to mark the equipment for completing the wells – in Table 4.

Table 3—Proppant with markers. Result of "blind" test to evaluate accuracy of markers identification in fluid samples

Ratio and codes identified by GEOSPLIT				Real ratio and codes mixed by the Customer №1					Discrepancy
Mixture	Cipher	Code	%	Mixture	Cipher	Code	%	Mass, g	%
1	WT	1	**24**	1	WT	1	**24,99**	249,95	**0,99**
	WG	2	**25**		WG	2	**25,34**	253,35	**0,34**
	WR	3	**16**		WR	3	**14,99**	149,98	**1,01**
	WU	4	**0**		WU	4	**0**	0	**-**
	WP	5	**35**		WP	5	**34,68**	346,88	**0,32**
	Total		**100**		Total		**100,00**	1000,16	**-**
2	AR	6	**29**	2	AR	6	**26,78**	273,76	**2,22**
	AQ	7	**18**		AQ	7	**18,16**	185,62	**0,16**
	AT	8	**11**		AT	8	**12,37**	126,47	**1,37**
	AY	9	**13**		AY	9	**12,32**	125,95	**0,68**
	AW	10	**29**		AW	10	**30,37**	310,5	**1,37**
	Total		**100**		Total		**100,00**	1022,3	-
Mean value of discrepancy, %									**0,94**

Table 4—Markers in granulate for cassettes. Result of "blind" test to evaluate accuracy of markers identification in fluid samples

Ratio and codes identified by GEOSPLIT			Real ratio and codes mixed by the Customer №2				Discrepancy
Mixture	Code	%	Mixture	Code	%	Mass, g	%
1	1	**31,00**	1	1	**27,03**	2,16	**3,97**
	2	**29,50**		2	**33,54**	2,68	**4,04**
	3	**-**		3	**-**	-	**-**
	4	**39,50**		4	**39,42**	3,15	**0,08**
	5	**-**		5	**-**	-	**-**
	Total	**100,00**		Total	**100,00**	7,99	**-**
2	1	**33,10**	2	1	**34,34**	5,78	**1,24**
	2	**20,20**		2	**20,32**	3,42	**0,12**
	3	**19,80**		3	**20,26**	3,41	**0,46**
	4	**-**		4	**-**	-	**-**
	5	**26,90**		5	**25,07**	4,22	**1,83**
	Total	**100,00**		Total	**100,00**	16,83	**-**
3	1	**8,20**	3	1	**9,27**	1,11	**1,07**
	2	**21,60**		2	**23,37**	2,80	**1,77**
	3	**42,10**		3	**31,47**	3,77	**10,63**
	4	**28,10**		4	**35,89**	4,30	**7,79**
	5	**-**		5	**-**	-	**-**
	Total	**100,00**		Total	**100,00**	11,98	**-**
4	1	**-**	4	1	**-**	-	**-**
	2	**27,50**		2	**31,13**	3,02	**3,63**
	3	**42,20**		3	**51,65**	5,01	**9,45**
	4	**-**		4	**-**	-	**-**
	5	**30,30**		5	**17,22**	1,67	**13,08**
	Total	**100,00**		Total	**100,00**	9,70	**-**
5	1	**32,10**	5	1	**38,27**	5,01	**6,17**
	2	**55,10**		2	**46,07**	6,03	**9,03**
	3	**-**		3	**-**	-	**-**
	4	**12,80**		4	**15,66**	2,05	**2,86**
	5	**-**		5	**-**	-	**-**
	Total	**100,00**		Total	**100,00**	13,09	**-**
Mean value of discrepancy, %							**4,54**

The results of the tests performed have determined:

1. In all sample mixtures, the signatures of the markers are correct 100% as determined;
2. The mean value of the discrepancy in testing with proppant was 0.94%, while testing with granulate − 4.54%.

Both tests were recognized as successful by the customers.

Conclusions

Testing is the most important stage in making a decision for customers about the application of this or that tracer technology. Comprehensive programs conducted by the provider of tracer technology helped in answering lots of questions and additionally made sure that the service company is capable of recognizing its own markers.

In our opinion, oil and gas producing companies should pay attention on the number of tracer technologies' tests conducted, which is an essential indicator of how well the technology has been worked out. At the same time, a mandatory type of testing must be an independent technology tests on the accuracy of the quantitative determination of tracers (markers), which can be claimed as a qualification requirement of the contractor during tendering.

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