Investigation of the Electrophysical Influence on the Antiwear Properties of Hydrocarbon Liquids

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The application of electrophysical influence on hydrocarbon liquids is considered in order to improve their performance and change the magnetic susceptibility of hydrocarbon fuels and oils. Experimental studies of the wear resistance of chromium ball-bearing steel ШХ-15 (ShKh-15) have shown that the application of the method of electrophysical influence to the absorption signal significantly improves the wear-resistance properties of fuel and reduces the wear of the contacting surfaces of steel in the environment of aviation kerosene TS-1 by 34-38 %. When exposed to the electrophysical method upon receipt of the emission signal (the occurrence of the effect of double nuclear resonance), the antiwear properties of kerosene are reduced to 85 %, which leads to an increase in wear of steel samples by 16-20 %. This is due to a decrease in the oxidative activity of the hydrocarbon medium by changing the interaction with paramagnetic oxygen and increasing the concentration of dissolved oxygen in the friction zone. The process of oxidation of steel goes to the formation of the final wear products with excess oxygen in the friction zone. Thick films of a mixture of oxides have low strength, they are brittle and are destroyed by friction. In the future, the oxidation process is repeated, intense wear and significant fluctuations in the friction force are observed, which leads to pronounced adhesion. Analysis of the results obtained by emission spectroscopy showed a certain relationship between the antiwear properties and the composition of kerosene TS-1. After the electrophysical exposure and run-in of kerosene, the content of resinous compounds quadrupled, the oxygenated compounds content decreased by ~ 23 % and sulphur compounds content decreased by ~ 25 %, which indicate the improvement of the antiwear properties of the kerosene and, consequently, the possibility of increasing the critical load. The electrophysical influence on the magnetic susceptibility of kerosene TS-1 was investigated. Experimental studies have shown an increase in kerosene magnetism. The maximum change of $\chi_d = -0.7227 \cdot 10^{-6}$ occurs 60-70 min after electrophysical exposure due to the relaxation processes characterizing the exchange of energy between the system of nuclear spins of hydrogen and electrons of paramagnetic centers (dissolved oxygen) with the formation of diamagnetic complexes.

Keywords: Hydrocarbon liquids, Magnetic field, Double nuclear resonance, Electrophysical method, Antiwear properties, Magnetic susceptibility.

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1. INTRODUCTION

At present, it is of great importance to increase the efficiency of hydrocarbon liquids use and the economic efficiency of vehicles power plants for the implementation of priority directions of energy saving.

The problems of efficiency and toxicity of vehicle power plants operation are directly related to the quality of used hydrocarbon liquids. In this regard, increasing the quality requirements and improving the essential operational properties of hydrocarbon liquids is a current and important scientific and technical question.

2. STATE OF THE PROBLEM

The effect of the magnetic field on hydrocarbon liquids is one way to improve their performance. Magnetic energy was used in researches [1-4] for the treatment of fuel, to reduce consumption, to increase thermal efficiency, as well as to reduce the emission of certain pollutants rates. In [5-7], the authors investigated the influence of an internal magnetic field on processes in tribocontact of textured dimple surfaces and the mechanism of wear under conditions of boundary friction with limited supply of lubricant. It is shown that along with traditional processes of formation and destruction of surface layer in tribocontact, there are processes of excitation of internal magnetic fields at edges of discrete regions. This results in the removal of wear products and a high rate of lubricant flow on regions of actual tribocontact in inter-dimple space, accelerating the process of regeneration of boundary lubricating film. Hydrocarbon liquids can be improved by external electrophysical effects on them [8, 9]. Thus, in [10], the results of studies of the change of heat transfer and mass in high-viscous hydrocarbon liquid by inductive and radio frequency electromagnetic heating are presented.

The basis of electrophysical influence is the interaction of molecular construction of a liquid system with external magnetic and electromagnetic fields [11]. The electrons of an atom and the nucleus of some elements have magnetic and mechanical moments. The spin of an electron appears as a thin structure in the atomic spectra. The magnetic moment of the electron interacts with the magnetic moment of the nucleus. The interaction energy depends on the mutual orientation of the spins or magnetic moments, and the number of possible orientations is determined by the spin of the nucleus. The

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total spin of the nucleus depends on whether or not the components of the nucleus compensate for each other.

In a magnetic field, the magnetic moments of the proton with the spin $I = \pm 1/2$ are oriented in only two ways – in the field or against the direction of the field. Each nucleus that has a magnetic moment, when it enters a magnetic field, acquires Zeeman's additional energy μH_0 [12].

As the external magnetic field strength increases, the energy difference between the spin directed against the field and the spin directed in the field increases, resulting in increased polarization. However, the spin formation does not occur immediately. Prior to the imposition of the magnetic field, the number of spins directed "up" and "down" was the same, and then in the field, the part of the spins directed against the magnetic field was inverted.

The nuclear spin reorientation is accompanied by a change in its energy, so it can only occur in the presence of some object with which the nucleus exchanges energy. Such objects may be electrons, adjacent nuclei, paramagnetic elements (O_2, etc.), ions, free radicals, or a high-quality radio frequency circuit tuned to the precession frequency of nuclei in an external magnetic field.

In the bulk of the substance there is a total magnetic moment of nuclei M (magnetization) directed along the outer field [12]

$$M = \frac{\mu_n^2 H_0 n_0}{kT},$$

where μ_n is the proton magnetic moment; H_0 is the magnetic field strength; n_0 is the number of nuclei per unit volume of the substance; k is the Boltzmann's constant; T is the temperature.

A sharp decrease in the magnetic field strength causes the extra energy of the proton to be absorbed by the atoms of the elements near the nuclei of hydrogen with a non-zero electron spin due to the superfine interaction. These elements can be paramagnetic oxygen, free radicals, etc., that is, paramagnetic centers interact with the basic chemical compound where these centers are placed.

Increase in the polarization of hydrogen nuclei by changing the magnetic field strength inevitably causes the excitation of electron shells in the molecules due to the superfine interaction. This causes induction currents that affect the physical properties of the liquids.

To improve the efficiency, stability and versatility of the magnetic effect on hydrocarbon liquids, an electrophysical method based on resonant energy absorption by the proton liquid system has been proposed. The method consists in simultaneously affecting the liquid of a non-uniform permanent magnetic field and a resonant high-frequency electromagnetic field H_1 [13]. The frequency of oscillation coincides with the frequency of precession of nuclei in a given magnetic field and there is a selective absorption of energy of the generator at the frequency H_0

$$\omega = \gamma H_0,$$

where ω is the frequency of nucleus precession; γ is the protomagnetic ratio of the proton.

The variable electromagnetic field H_1 causes excitation of the nuclear spin system, which is described by the Hamiltonian. The influence of the electromagnetic field causes intense "bottom-up" transitions and vice versa, between the levels and the Boltzmann distribution population is disturbed. In this case, the field electromagnetic energy is partially absorbed by the proton liquid system, the absorption signal is observed, and, therefore, the energy of the medium is increased [14].

Therefore, the resonant absorption of the oscillatory circuit electromagnetic energy by the proton liquid system and the exchange of this energy with the liquid molecular system lead to changes in the physical and chemical parameters of the hydrocarbon liquids. Ultimately, the electrophysical effect leads to a change in the rate constant of the chemical reaction, which is determined by the Arrhenius law, by changing the activation energy, which in turn affects the speed and completeness of combustion and as a result, the performance of vehicles [2].

Thus, the considered theoretical aspects of the magnetic influence on the hydrocarbon liquid systems prove the change in the energy of interaction of the nucleus magnetic moment with the spin electron system. Depending on the method of influence, namely, the receipt of the absorption signal in inhomogeneous magnetic fields or the emission signal using the effect of double nuclear resonance, it is possible to manage the proton hydrocarbon liquid system by the electrophysical method of influence. This allows increasing and decreasing the energy state of the hydrocarbon liquid and thereby affecting the physicochemical and operational properties of the hydrocarbon liquids.

The purpose of the work is to increase the physicochemical and operational properties of hydrocarbon liquids by electrophysical method of influence on the proton system to obtain the effect of double nuclear resonance.

3. RESEARCH RESULTS AND THEIR DISCUSSION

The special device (Fig. 1) was developed to determine changes in the qualitative characteristics of hydrocarbon liquids after electrophysical influence. The device consists of a permanent magnet 1. In the field of the permanent magnet, there is a polarizing volume 2, from which the liquid through pipeline 3 enters the inhomogeneous scattering fields of the permanent magnet 1. In the scattering fields of this magnet, there is a high-frequency coil 4 of the oscillating LC-circuit, which is connected to a high-frequency generator 5.

Experimental studies under sliding friction condition were performed in the environment of a carbohydrate liquid, which is selected as the kerosene TS-1. The material of the friction couple was chromium ball-bearing steel ШХ-15 (ShKh-15). Antiwear properties of kerosene (fuel) were investigated under three types of exposure. In the first case, the fuel was exposed to the magnetic field of two permanent magnets arranged in series with the magnetic field strength of 340 kA/m and 156 kA/m, respectively. The fuel flow rate was determined by the maximum absorption signal, after which high and low frequency generators were switched off.

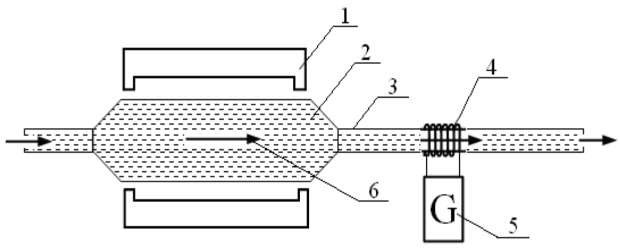

Fig. 1 – Diagram of the device for electrophysical influence on hydrocarbon liquids

In the other two cases, the fuel was affected by the electrophysical method, that is, the effect on the fuel occurred in the constant magnetic fields with the generators turned on. In this case, the absorption of the electromagnetic energy of the *LC* oscillatory circuit was absorbed by the system of nuclear spins of hydrogen, i.e., the inversion took place and the absorption signal was observed. The fuel flow rate was controlled by the maximum absorption signal.

In the third case, in order to obtain a double nuclear resonance under the electrophysical influence on the proton system of the hydrocarbon liquid, an additional radio frequency coil of nutation was installed between the polarized magnet and the high frequency coil of the absorption signal, which was connected to the sinusoidal oscillator. As a result, they observed an emission signal, which meant the return of Zeeman energy by a system of nuclear spins of hydrogen oscillating circuit.

The nutation coil was positioned in a non-uniform magnetic field and oriented so that the liquid flowed along the gradient and the oscillatory field lines were directed perpendicular to the external magnetic field. The formation of the longitudinal component of nuclear magnetization results from the exchange of energy between the nuclear spin system and the molecular system. The rate of this exchange, and therefore the formation of the longitudinal component, is determined by the intensity of the spin-lattice interaction.

From the difference of population levels, it follows that the substance has a magnetic susceptibility equal to

$$\chi = \chi' - j\chi'',$$

where χ' is the complex susceptibility of the sample; $j\chi''$ is the losses in determining the magnetic susceptibility that determine the absorption of the power of an alternating electromagnetic field per unit volume of the sample [15]. The relationship between microscopic and macroscopic properties can be established when calculating the average value of the energy P_ω of alternating electromagnetic field $H_1\cos\omega t$, which is absorbed per unit time,

$$P_\omega = 2\omega\chi'' H_1^2 \cos\omega t.$$

This equation establishes a simple relationship between the absorbed power P, the susceptibility χ'', and the amplitude of the alternating magnetic field H_1.

The value increases sharply near the resonance and causes a strong resonance absorption of energy from the oscillating circuit of the high-frequency generator. In this case, there is a signal of energy absorption, that is, the energy absorbed by the nucleus (ΔE) during the transition from one energy level to another.

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$$\Delta E = h\gamma H_0,$$

where h is the Planck constant; γ is the alternating electromagnetic field.

Investigation of the electrophysical influence on the magnetic susceptibility of the fuel TS-1 was carried out on the radio frequency installation whose principle of action is based on the determination of the change in the resonant frequency of the high-frequency oscillation circuit. In the measurement process, the accuracy of the frequency stability was up to 10^{-8}, which allowed us to determine the diamagnetic susceptibility of fuels of the order of 10^{-6} to within 0.01 %.

4. RESULTS AND DISCUSSION

The analysis of experimental studies allowed us to establish that the influence on the fuel of a permanent magnetic field leads to a reduction of wear of steel ShKh-15 at sliding friction in the environment of kerosene TS-1 by 10-14 %, which corresponds to the index of antiwear properties $K = 116$ % (Fig. 2). The application of the method of electrophysical influence at the absorption signal significantly improves the wear-resistance properties of the fuel and reduces the wear of the contacting surfaces of steel in the medium of TS-1 by 34-38 % ($K_a = 164$ %). Under the influence of the electrophysical method, but upon receipt of the emission signal, the antiwear properties of the fuel deteriorate, which leads to an increase in wear of steel samples by 16-20 %. The index of the antiwear properties of the fuel decreases to equal $K_e = 85$ %. It is found that the decrease in the antiwear properties of the fuel leads to the adhesion of 30 % of friction samples in the study of the effect of nutation (double nucleus resonance – emission signal).

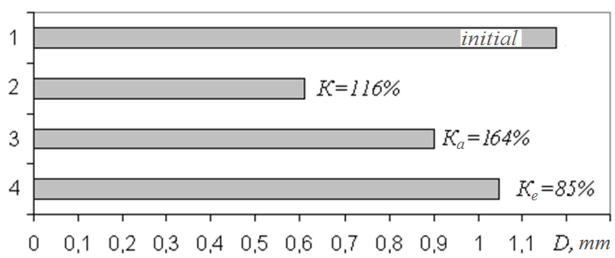

Fig. 2 – Change of the antiwear properties of steel ShKh-15 depending on the electrophysical influence in the environment of kerosene TS-1: 1 – initial sample; 2 – after the exposure of a constant magnetic field; 3 – after the electrophysical exposure (absorption signal); 4 – after the electrophysical exposure (emission signal)

The studies of fuel TS-1 by emission spectroscopy showed no iron in the original fuel. In the fuel sample after run-in, but without the electrophysical exposure, iron of 0.00048 % by weight was found, and in the fuel sample, which was subjected to electrophysical exposure the iron content was twice less – 0.00024 %. In addition, the analysis of the obtained results showed a certain relationship between the antiwear properties and the composition of the fuel. After electrophysical exposure and fuel run-in, the content of the resinous compounds quadrupled, the content of oxygenated compounds decreased by ~ 23 %, and the content of sulfur compounds decreased by ~ 25 % (Table 1). All this in the complex speaks about improvement of the antiwear properties of fuel, and therefore about the possibility of increasing the critical load.

Table 1 – Changes in the chemical composition of fuel TS-1 before and after testing

Name of indicators	Original fuel, %	Fuel after run-in, %	
		without electrophysical exposure	after the electrophysical exposure
Content of resinous compounds	–	0.006	0.024
Sulfur content	0.45	0.37	0.27
Content of oxygenated compounds	0.075	0.35	0.27
Content of nitrogenous compounds	1.2	1	1.1

Hydrocarbon fluids are carriers of natural additives (molecular oxygen) that are actively involved in the process of boundary friction. Therefore, the formation of oxide films in the friction zone occurs in the process of oxidation of metal and hydrocarbons. Three factors are important: the oxidative activity of the medium, the oxidation of hydrocarbons and the conditions for the transfer of molecular oxygen to the friction zones. Therefore, the wear of friction pair decreases due to the decrease in concentration of dissolved oxygen spent on the oxidation of liquid medium and the boundary films formation from products of oxidation.

Thus, the results of the antiwear properties studying of fuel TS-1 show that the electrophysical influence with the receipt of an absorption signal leads to a decrease in the amount of wear of steel ShKh-15, and therefore, to an improvement of the antiwear properties of fuels.

The increase in wear after electrophysical exposure with the receipt of an emission signal (double nuclear resonance) is due to a decrease in the oxidative activity of the hydrocarbon medium by changing the interaction with paramagnetic oxygen and increasing the concentration of dissolved oxygen in the friction zone. In other words, with excess oxygen in the friction zone, the oxidation process of metals leads to the formation of the final products. Thick films of a mixture of oxides have low strength; they are fragile and are destroyed by friction forces. In the future, the oxidation process is repeated and intensive wear and significant fluctuations in the friction force are observed. In low molecular weight hydrocarbon liquids, excess oxygen in the friction zone results in a pronounced adhesion (up to 30 % of tests).

Hydrocarbon fuels can be represented as a diamagnetic chemical compound with paramagnetic centers representing organic radicals, dissolved oxygen, and the like. Hydrocarbon molecules of the fuel do not contain unpaired electrons and their magnetic properties are due to the diamagnetism of closed electron shells and the weak paramagnetism of atomic nuclei (hydrogen nucleus) [14].

When the diamagnetic substance is introduced into the magnetic field, the electron shell energy changes by ΔU, which leads to the appearance of the magnetic moment of the shell μ_z and the magnetization of the substance

$$M_z^d = -\frac{e}{2mC} N \sum_{i=1}^{z} < L_z > - \frac{Ne^2}{6mC^2} H_0 \sum_{i=1}^{z} < r_i^2,$$

where N is the number of atoms of the substance; e is the charge of the electron; m is the mass of the electron; C is the light speed.

The first part of the formula is related to the orientation of the orbital moments of atoms in the absence of an external field and in our case it is zero. The second part of this expression is always negative and defines the magnetic part of the magnetization

$$M_z^d = \chi_0^d H_0,$$

where χ_o^d is the diamagnetic susceptibility,

$$\chi_0^d = -\frac{Ne^2}{6mC^2} \sum_{i=1}^{z} < r_i^2.$$

Diamagnetic susceptibility is very small and for condensed media is

$$\chi_0^d = -\left(10^{-5} - 10^{-6}\right).$$

The magnetic moments of such particles as oxygen, organic radicals and other fuel elements having electronic paramagnetism are oriented under the action of the external magnetic field and their magnetization is proportional to the external magnetic field H_0

$$M_z^p = \chi_0^p H_0.$$

Because the moments are oriented in the direction of the field, so $\chi_0^p > 0$

For the effect we consider, the magnetization of the substance in which there are nuclear moments and there is a subtle interaction of the nuclear moments with the magnetic moments of unpaired electrons is due to the total magnetic moments of atoms and molecules in the excited state. The interaction of hydrogen nuclear spins, which received alternating electromagnetic field energy during the electrophysical influence, with electron spins of paramagnetic centers due to electron-nuclear spin interaction leads to a change in the magnetization of the substance. The interaction of the electrons spins of the paramagnetic centers with the spins of the nuclei of the basic chemical compound where these centers are placed leads to an increase in the substance's magnetism, i.e., the electron sharing of each of the atoms will occur, leading to a decrease in the dissolved oxygen paramagnetism.

Of considerable interest is the study of the effect of electrophysical influence on the magnetic susceptibility of hydrocarbon liquids. Experimental studies of the magnetic susceptibility of fuel TS-1 showed an increase in fuel diamagnetism (Fig. 3).

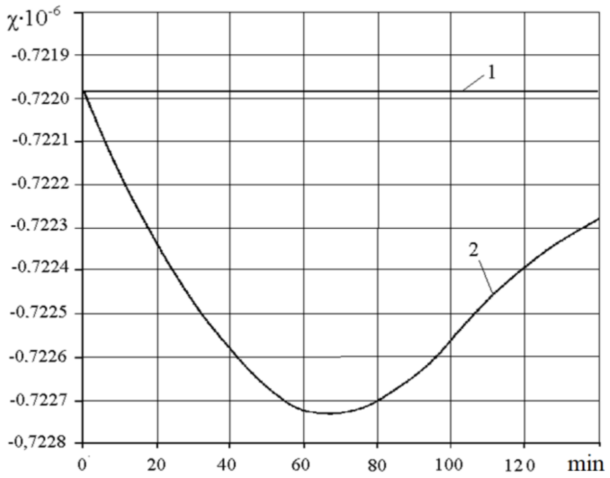

Fig. 3 – Change of magnetic susceptibility of fuel TS-1 depending on time: 1 – the source fuel; 2 – the fuel after electrophysical influence

It was found that the maximum change in $\chi^d = -0.7227\cdot10^{-6}$ occurs 60-70 min after electrophysical influence. This is probably due to the relaxation processes that characterize the exchange of energy between the hydrogen nuclear spin system and the electrons of the paramagnetic centers (dissolved oxygen), forming diamagnetic complexes.

The liquid, which has been in a constant magnetic field for a long time, has a uniform magnetization of nuclei directed parallel to the field. A slow change in the magnetic field direction causes the nuclei to rotate with it. If the rotation of the magnetic field takes place at a time t much smaller than the precession period of the nuclei, then the nuclei magnetization does not have time to return and deviates from the magnetic field direction. This effect was first discovered by Purcell and Pound [16].

The deviation of the nuclei magnetization from the magnetic field direction in the flowing liquid, which was previously polarized in a strong magnetic field, was due to the flow of liquid inside the nutation coil. The nutation coil is in inhomogeneous magnetic fields. Then the liquid was fed into the coil of high frequency exposure and the amplitude of the absorption signal varied at the output of the device.

The influence of the oscillating electromagnetic field with a frequency equal to the precession frequency of the nuclei in a given inhomogeneous magnetic field, deviates the magnetization of the nuclei from the field direction by some angle (nutation angle), which is determined by the expression

$$\theta = \gamma H_1 \Delta t,$$

where H_1 is the intensity of the alternating electromagnetic field, A/m.

The magnetization of nuclei M in the liquid flowing into the high-frequency coil is proportional to the projection of the nuclei magnetization in the liquid flowing from the nutation coil and depends on the time of liquid motion between the coils

$$M = M_{z\,out} e^{-V_T / qT_1}.$$

The magnitude of M is proportional to the amplitude of the absorption (emission) signal in the high frequency coil.

Under resonant conditions, the energy of the electromagnetic field is absorbed in the coil of nutation, which leads to the transition of nuclear spins from the lower energy level to the upper one. If liquid that has an energy level population higher in the upper state than in the lower state enters the high frequency coil (standing after the nutation coil), then the resulting absorption will be negative, that is, the system gives off more energy than it receives. This changes the absorption signal in amplitude and sign.

Thus, the excitation in the oscillation sensor of the oscillating field at setting the resonant frequency to the maximum of the effect of nutation and selection of the oscillation amplitude on the nutation coil in the high frequency oscillation circuit appears a negative absorption signal. This means that when the negatively polarized liquid flows into the high-frequency coil, an emission signal is observed in it instead of the absorption signal. That is, the spin system of the nuclei emits energy into the radio frequency oscillation circuit and an emission signal is observed, which indicates a decrease in the energy state of the proton system and the liquid.

5. CONCLUSIONS

Studies of the wear resistance of steel ShKh-15 in the environment of kerosene TS-1 have shown an improvement in the antiwear properties of the kerosene and the wear resistance of the contacting surfaces of the steel when applying the method of electrophysical exposure at the absorption signal. In the case of the effect of double nuclear resonance (upon receipt of the emission signal), the antiwear properties of the kerosene deteriorate, which leads to a decrease in the wear resistance of steel ShKh-15 by reducing the oxidizing activity of the hydrocarbon medium and increasing the concentration of dissolved oxygen in the friction zone.

Studies have shown an increase in the kerosene TS-1 magnetism after electrophysical exposure due to relaxation processes that characterize the energy exchange between the hydrogen nuclear spin system and the electrons of paramagnetic centers with the formation of diamagnetic complexes.

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Дослідження електрофізичного впливу на протизносні властивості вуглеводневих рідин

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Розглянуто застосування електрофізичного впливу на вуглеводневі рідини з метою поліпшення їх експлуатаційних показників та зміну магнітної сприйнятливості вуглеводневих палив і олив. Експериментальними дослідженнями зносостійкості сталі ШХ-15 встановлено, що застосування способу електрофізичного впливу при сигналі абсорбції значно покращує протизносні властивості гасу і зменшує знос контактуючих поверхонь сталі в середовищі гасу ТС-1 на 34-38 %. При впливі електрофізичного способу при отриманні сигналу емісії (виникнення ефекту подвійного ядерного резонансу) протизносні властивості гасу погіршуються, що призводить до збільшення зносу зразків сталі на 16-20 %. Показник протизносних властивостей гасу при цьому зменшується до 85 %. Це пояснюється зменшенням окислювальної активності вуглеводневого середовища за рахунок зміни взаємодії з парамагнітним киснем і збільшенням концентрації розчиненого кисню в зоні тертя. При надлишку кисню в зоні тертя процес окислення сталі йде до утворення кінцевих продуктів зношування. Товсті плівки суміші окислів мають невисоку міцність, вони крихкі і руйнуються під дією сил тертя. В подальшому процес окислення повторюється і спостерігається інтенсивний знос і значні коливання сили тертя, що призводить до яскраво вираженого схоплювання. Аналіз отриманих результатів методом емісійної спектроскопії показав певний зв'язок між протизносними властивостями і складом гасу ТС-1. Після електрофізичного впливу і напрацювання гасу в чотири рази збільшився вміст смолистих сполук, на ~ 23 % зменшився вміст кисневих сполук і на ~ 25 % сірчистих, що говорить про поліпшення протизносних властивостей гасу, а отже і про можливість збільшення критичного навантаження. Досліджено електрофізичний вплив на магнітну сприйнятливість гасу ТС-1. Експериментальними дослідженнями встановлено збільшення діамагнетизму гасу. Максимальна зміна $\chi^d = -0.7227 \cdot 10^{-6}$ відбувається через 60-70 хвилин після електрофізичного впливу за рахунок релаксаційних процесів, що характеризують обмін енергією між системою ядерних спінів водню і електронами парамагнітних центрів (розчиненого кисню) з утворенням діамагнітних комплексів.

Ключові слова: Вуглеводнева рідина, Магнітне поле, Подвійний ядерний резонанс, Електрофізичний спосіб, Протизносні властивості, Магнітна сприйнятливість.