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Influence of physico-chemical parameters of surface-active systems components for minimization of evaporation of hydrocarbon liquids

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Highly efficient stable aerated hydrophilic compositions containing fluorotensides and ultralight microdisperse systems using gas-filled glass, aluminosilicate and polymer microspheres have been developed. Designing the compositions of PAS based on the surface activity of surfactants, their solubility in water and the ability to bind water and the formation of hydrogen bonds between the components. The main condition for the stability of the coating when mixing the components - the chemical interaction between them and the formation of a system that does not dissolve in hydrocarbons and does not break down in terms of use. The best film-forming characteristics necessary for the operation of the coating (simultaneous reduction of surface tension and film formation) active substances (FPAR), the non-polar part of the molecules of which contains a fluorocarbon chain, so they are insoluble in hydrocarbons, well soluble in water and easily distributed on the surface of hydrocarbon liquids, creating a protective film. The choice of co-surfactants was based on the ability to stabilize hydrophilic films on the surface of hydrocarbons not only at favorable HLB, but also at the lowest, although higher than the critical concentration of micelle formation (CCM), concentrations for forming a mixed adsorption layer of increased strength. This surfactant was water-oil-soluble twin-80, which will significantly increase the hydrophilic part and enhance the stability of the PAS and the stability of the aerated system. The introduction of glass microspheres into the components of the system has significantly enhanced its strength and stability.

In the study of the stability and gas permeability of the developed surfactant systems, it was found that the insulating ability of the coating increases with increasing hydrophilic-lipophilic balance of the system and due to chemical interaction between the carboxyl group of fluorotenside and hydroxyl groups of surfactants.

Keywords: evaporation of hydrocarbon liquids, stability of surface-active system, hydrophilic-lipophilic balance, glass microspheres

Introduction

During storage of the liquid fuels, a significant part of them is lost (80% of all losses) because of evaporation. For example, for the tank with a volume of 5000 m^3, about the 4 t of gasoline are lost, during 1 year these losses may increase up to 230 t. Improvement of the fuel storage is one of the most important problems. Its solution will allow not only to save the fuel quantitative and qualitative composition, but also to prevent complex of emergencies at tankfarms [1,2].

Petroleum products are known to be a multicomponent mixture of hydrocarbon compounds having different boiling points. The lightest of them are automobile (boiling point - 35-40 ^oC) and aviation gasoline (boiling point – above 40 ^oC). During storage, the quality of gasoline deteriorates, mainly due to the evaporation of light fractions of hydrocarbons C_5-C_7, which are the most volatile [3]. This leads to an increase of the boiling point and an increase of density due to the weighting of the fractional composition of the fuel.

Although the problem of loss of liquid hydrocarbon fuels during storage has been worked on for a long time, it is still relevant, because of the low research of the processes to prevent evaporation and to prevent the occurrence of explosive concentrations of hydrocarbon liquids [4], as well as the lack of effective technological systems and tools to minimize them. Solving the problem of preventing evaporation of liquid hydrocarbon fuels by targeted production of surface-active systems (SAS) with certain protective properties is relevant in general scientific terms, and the creation of technological aspects of their use is extremely important in practice: environmental and economical.

The goal of the work. The aim of the work is to develop an effective protective coating to prevent evaporation of hydrocarbon liquids, to increase the safety of their storage and protect the environment. To detect the relationship between the structure and insulating ability of surfactant molecules, to research of stability and gas permeability, the insulating ability of the developed surfactant systems when comparing the evaporation of individual hydrocarbon liquids C_5-C_{10}, model mixture and technical gasoline A-95 from the open surface and when applied to the surface of the ultralight surfactant system with the inclusion of glass microspheres.

Materials and methods

Industrial unfinished and finished microspheres with a size of 25 μm with a bulk density of 0.30 g / cm^3 were used in the work.

Unfinished microspheres are white, thin-walled spheres made of sodium borosilicate glass according to TU 6-48-108-94 type MS group A1 with a density of 240 g / cm^3 and a hydrostatic compressive strength of 8 MPa.

Finished microspheres are glass beads with a mixture of gamma- and beta-aminoisopropyltriethoxy-silane (product AGM-9; TU 6-02-724-77) type MS-A9 group A1 applied to their surface (0.3%) Fig.1.

Fig.1. Microphotography of the surface of the hydrocarbon liquid partially covered with microspheres.

Surfactants (surfactants): nonionic surfactant sorbitan monooleate polyethylene glycol, fluorinated surfactant 2- (N-ethylperfluorooctansulfamide) acetic acid, polymer-polyvinyl alcohol (PVA), stabilizer. The experiment was carried out as follows: at the same time studied the evaporation by analytical method, placed in a thermostat of cylindrical tanks with a volume of 100 ml at a temperature of 25 ^{o}C model samples of volatile individual hydrocarbon liquids C_5-C_{10} (gasoline components) from an open surface of 20 cm^2 microspheres on the same surface with a monolayer and a layer that exceeded the surface area of hydrocarbons twice or three times. The studies were performed under identical conditions for 5 h, weighing the samples every 10 min. Under the same conditions, the evaporation of hydrocarbon liquids during the application of an ultralight surfactant system was investigated.

The study of the effectiveness of the developed ultralight surfactant systems (USAS) to prevent evaporation of the model mixture, simulating the composition of gasoline, and gasoline A-95 was performed on an experimental setup using the chromatographic method of analysis. Hydrocarbons invert microdispersions were prepared by gradually adding an aerated hydrophilic solution of FPAR (fluorotensides), tween-80, and PVA to a pre-prepared solution of basic surfactant (emultal, oleodin, phosphatidine) in a hydrocarbon solvent, followed by stirring for 10 min.

To obtain emulsion-suspension systems, gas-filled microspheres were introduced into the finished highly structured inverse emulsion with constant stirring for 7-10 min. According to previous studies, this time was sufficient to establish the equilibrium adsorption of surfactant molecules and obtain a stable emulsion-suspension composition.

Research results: highly effective stable aerated hydrophilic fluorotensin-containing compositions and ultralight microdisperse systems using gas-filled glass, aluminosilicate and polymer microspheres were created.

Discussion of results. When designing SAS compositions, were used the surface activity of surfactants, their solubility in water and the ability to bind water and the formation of hydrogen bonds between the components. The main condition for the stability of the coating when mixing the components is the chemical interaction between them and the formation of a system that does not dissolve in hydrocarbons and does not break down in use. Based on the above stated, the priority was given to surfactants of the anionic type, containing in the alkyl chain of 6-12 carbon atoms, which are available at the market and have a high surface activity. Among our surfactants tested, the best film-forming characteristics required for the coating to function (simultaneous reduction of surface tension and film formation) showed fluorinated surfactants (FPAR), the non-polar part of the molecules of which contains a fluorocarbon chain, so they are insoluble in carbohydrates, but are well soluble in water and are easily distributed on the surface of hydrocarbon liquids, creating a protective film. Perfluorocarboxylic and perfluoroalkane-sulfonic acids containing 6-12 carbon atoms in the fluorinated chain were used. The hydrophobic chain of such compounds is immersed in liquid hydrocarbons, and hydrophilic groups of four S-N-C-C atoms with a functional carboxyl group are concentrated on the interface with air. Air was pumped through aqueous solutions to ensure buoyancy.

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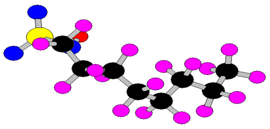
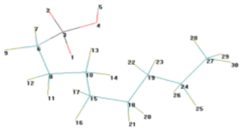
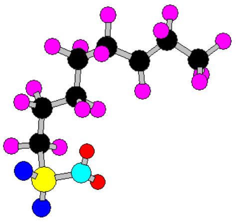
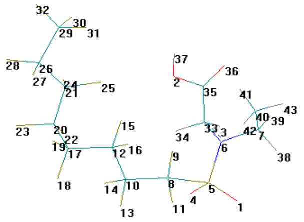




$C_8F_{14}SO_3H$
$R_fSO_2NH_2$





When applying such an aerated system based on AFFF-3M foaming agent on the surface of gasoline, the insulating capacity of the foam, as expected, increases, but the integrity and stability of the surface structure and, accordingly, protective properties are lost after a short time [6]. Therefore, there was a need to stabilize it, increasing the hydrophilic-lipophilic balance of the system by connecting other molecules with a large number of hydrophilic groups.

The choice of co-surfactants was based on the ability to stabilize hydrophilic films on the surface of hydrocarbons not only at favorable HLB, but also at the lowest, although higher than the critical concentration of micelle formation (CCM), concentrations for forming a mixed adsorption-MES layer on the surface strength. This surfactant was water-soluble twin-80 [8]. This nonionic surfactant does not dissociate into ions, is hydrophilic, oil-soluble and insoluble in hydrocarbons. The molecular structure of the compound includes a tetrahydrofuran ring with three hydrocarbon chains containing a carbonyl group, 20 oxymethylene groups and two hydroxyl groups. After studying the structure of tween-80, it is assumed that when mixed with FPAR due to the chemical interaction of tween-80 on hydroxyl groups with carboxyl groups FPAR esters are formed, which will significantly increase the hydrophilic part and enhance the stability of SAS and the stability of the aerated system.HLB = (4 + 15) / 2 = 9.5 Stability - 3 hours.

However, the stability of such a layer does not meet the stated requirements. In fact, if the half-life of the aerated system formed by fluortenside AFFF-3M does not exceed 8 minutes, then in conjunction with tween-80 a satisfactory protective layer of foam (at least 10 mm) is stable for more than 3 hours. However, these results are insufficient for practical use.

It is established that the only way to ensure long-term stability of aerated systems is the formation of three-phase solid systems [6]. Typically, such systems are obtained by introducing into the composite composition of the surfactant polymeric structurant. Combinations of surfactants with polymers are used for many practical purposes, which provides stability and the necessary rheological properties of the compositions [8]. The properties of the solutions of the individual components are important, but the performance of the composite product largely depends on the interaction between the components. That is, knowledge of physicochemical properties of components and features of interaction between them is essential not only for the correct combination of the best properties of components, but also for creation of composite systems with fundamentally new properties, which is a combination of science and art [7]. It was found that the stretching of the surfactant polymer increases the stability of the coating. When introducing a polymer into the coating composition, it should be borne in mind that the viscosity of polymer solutions is usually higher than the viscosity of solutions of low molecular weight compounds with the same concentrations. The swelling process is a diffusion process caused by the gradient of the chemical potential of the liquid.The cause of swelling is that molecules that differ by many orders of magnitude in size and mobility mix and interact. Low molecular weight solvent molecules rapidly infiltrate the polymer structure, pushing the chains apart and increasing the volume. From this point of view, swelling is a thermodynamic process with a unilateral equilibrium shift, in which the solvation of its macromolecules occurs (polar polymers swell in a polar solvent), accompanied by heat release, ie $\Delta H < 0$. At this stage, the entropy of the system does not change or decreases slightly due to the ordering of solvent molecules due to solvation, ie $\Delta S < 0$. If $\Delta H < T\Delta S$, then $G < 0$ is an spontaneous process. The amount of solvent absorbed at this stage is 20-40% by weight of dry matter, and despite the increase in the volume of the polymer sample, the total volume of the system (polymer and solvent) at this stage decreases, i.e. there is a contraction and the interaction of polymer with solvent – solvation, which results in compaction of the system. In this case, the spatial grid is created not by vanderwaals, but by chemical or hydrogen bonds. For this reason, gellies are not characterized by thixotropy and destruction of the structure of gels due to mechanical impact, which in most cases is irreversible. Gels formed by polymers exhibit elastic and resilient properties due to the strength and flexibility of the macromolecular network, as well as the hardness of the oriented layers of solvent molecules. For this reason, conventional gels are not capable of plastic flow, while globular gels are able to flow. The mechanical properties of gels are strongly influenced by their concentration. Due to the compaction of gels, their strength increases.

Given the described set of general properties of polymers, after a careful search among high-molecular water-soluble surfactants: natural – cherry gum, gelatin, egg white, and synthetic - polyacrylamide, copolymers of maleic anhydride and, we focused on polyvinyl alcohol, which is synthesized exclusively by the method of polymeranalogical transformations. The matrix polymer is polyvinyl acetate, the ester groups of which are subjected to hydrolysis.

X-ray films of PVA become insoluble even in boiling water and can withstand heating up to 200 °C. Elongated films, despite the presence of plasticizer, have increased strength (tensile strength - 600 kgf / cm) and high abrasion resistance. The gas permeability of PVA films is 15-20 times (depending on the degree of plasticization) is lower than the gas permeability of the vulcanized natural rubber film and is a consequence of the orientation of macromolecules due to numerous hydrogen bonds between neighboring macromolecules. The combination of a polymer matrix PVA with many centers that can form hydrogen bonds, with an aerated solution of SAS allowed to develop compositions of stable three-phase systems that form on the surface of hydrocarbons strong three-dimensional elastic systems that provide gas-tight coatings and provide reliable liquids (Fig. 2).

The formed complex forms on the surface of hydrocarbons structured elastic films. The great practical value of PVA in a peculiar combination of a number of advantageous properties. Due to the high gas impermeability of this thermoplastic microcrystalline structure, its hydrophilicity, solubility in water and at the same time insolubility in hydrocarbons, on the basis of previously created SAS we were able to develop durable coatings to protect hydrocarbon liquids from evaporation systems in combination with surfactants are glass microspheres [8].

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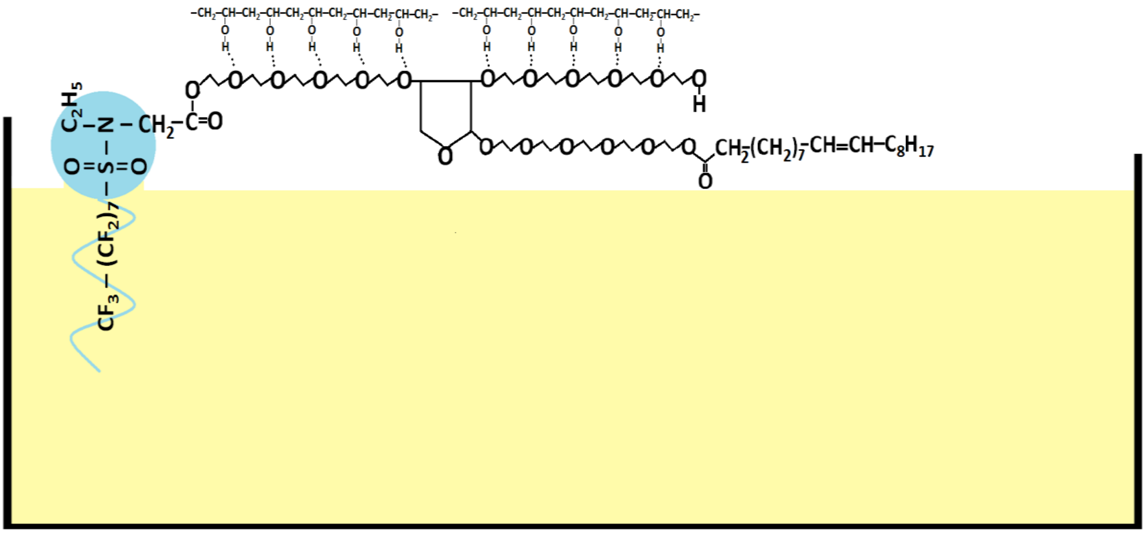

Fig. 2. Scheme of the coating formed by the interaction of 2- (N-ethylperfluorooctane sulfamide) acetic acid, sorbitan monooleate, polyethylene glycol and polyvinyl alcohol.

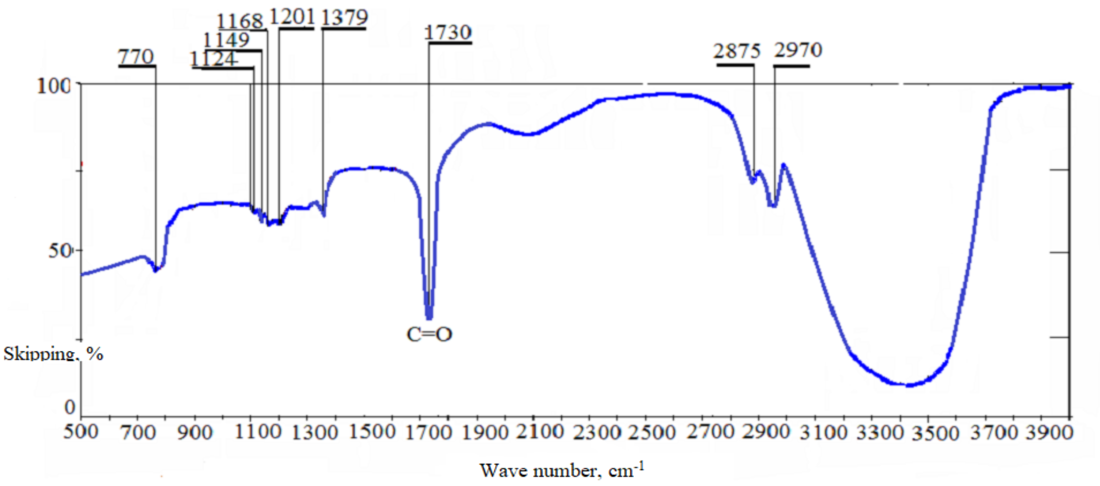

Fig.3. IR spectrum of 2-(N-ethylperfluorooctansulfamide) acetate.

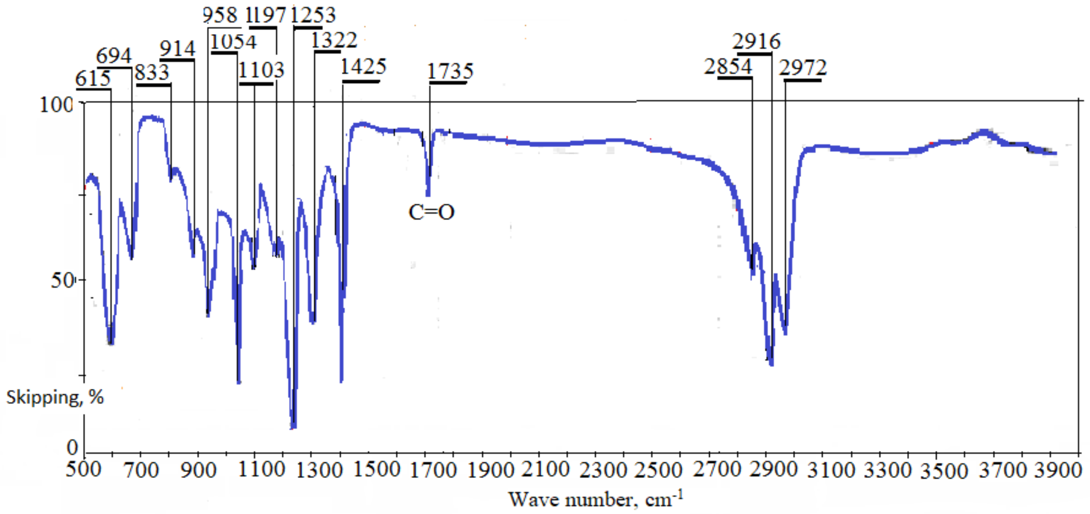

Fig.4. IR -spectrum of polyethylene glycol sorbitan monooleate.

The IR-spectrum of 2-(N-ethylperfluorooctansulfamide) acetate is shown in Fig.3. The hydrophobic part of this molecule includes one fluoromethyl and 7 fluoromethylene groups. In the spectrum they correspond to the absorption bands of deformation oscillations CF_2, CF_3 groups in the range of 1168 cm^{-1}, 1149 cm^{-1}, respectively. The hydrophilic part of the molecule consists of four S-N-C-C atoms with a functional carboxyl group, which includes a carbonyl group C = O with a corresponding absorption band of 1730 cm^{-1}, and a hydroxyl group characterized by a wide absorption band in the range of 3100-3700 cm^{-1}.

To approximate the oscillation frequency of the valence bond according to Hooke's law, the relative contributions of the bond strength and atomic masses must be considered. For example, a superficial comparison of a CH group with an F-H group based on atomic masses may lead to the conclusion that the F-H valence frequency should occur at a lower frequency than for the C-H bond. However, the increase in the force constant from left to right in the first two periods of the periodic table has a greater effect than the increase in mass. Thus, the group F-H absorbs at a higher frequency (4138 cm^{-1}) than the C-H group (3040 cm^{-1}), because the energy of the chemical bond C-F significantly exceeds the energy of the bond C-H. In general, functional groups that have a strong dipole give strong absorption in the IR-spectrum.

Figure 4 shows the IR-spectrum of tween-80, which shows the oscillations of functional groups and their bonds, in particular, the strong absorption band of valence oscillations of the C=O bond at 1735 cm^{-1}. This band is very sensitive to changes in the physical state of the compound. The highest value of frequencies $\nu_{C=O}$ is observed for compounds in pairs, then for solutions of carbonyl compounds in nonpolar or low-polar solvents (CS_2, CCl_4) and, finally, even less value of frequencies $\nu_{C=O}$ acquires in solutions of polar solvents ($CHC1_3$, $CHBr_3$). The lowest values of the frequencies of the carbonyl group are observed in the solid state, where intermolecular interactions play a significant role. If the carbonyl group is involved in the formation of intermolecular hydrogen bonds, the frequency $\nu_{C=O}$ decreases to 1545 cm^{-1}.

The intermediate part of the spectrum in the range of 1300-900 cm^{-1}, is usually referred to as the range of «fingerprints». The absorption of the sample in this area is often complex, with bands of oscillations in the interaction of oscillatory modes. This part of the spectrum is extremely valuable in the study with reference to other ranges [9].

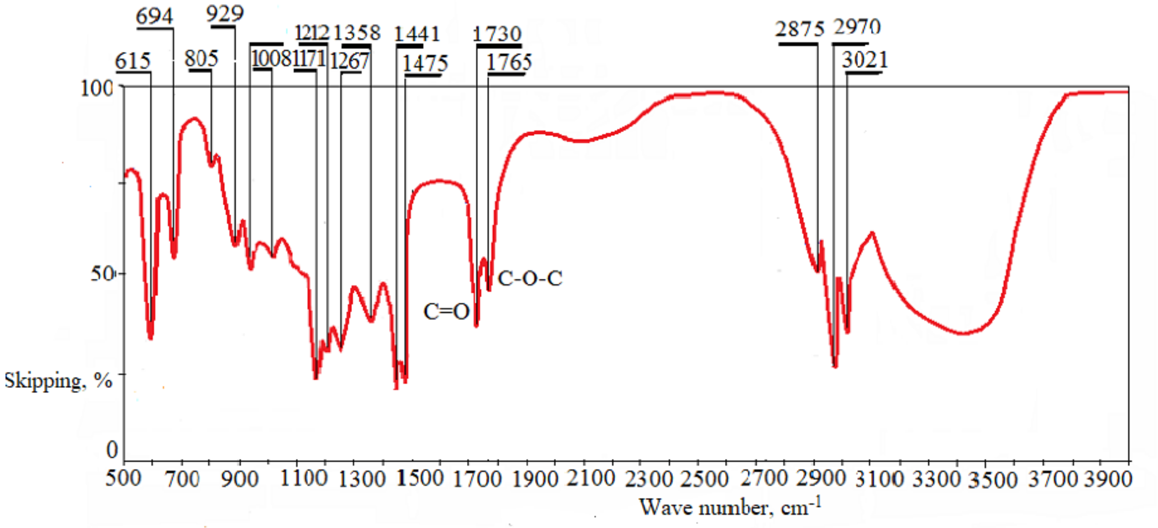

Fig.5. IR spectrum of a mixture of 2- (N-ethylperfluorooctansulfamide) acetic acid and polyethylene glycol sorbitan monooleate.

The assignment of a carbonyl group in the ester is confirmed by the observation of a strong band in the region of 1300-1100 cm^{-1}, which indicates a CO-valence oscillation.

The HO-valence oscillation ν_{OH}, one of the most characteristic of the IR spectra, forms the most intense band in the range of 3200-3600 cm^{-1}. The position and nature of the band also depend on the degree of participation of the hydroxyl group in the hydrogen bond. The hydrogen bond changes the force constant of the bond -OH and, accordingly, leads to a decrease in wave oscillations, which is traced in the spectrum after the introduction into the SAS of a polymeric structurant - polyvinyl alcohol. As can be seen from Fig.3.6, in a pure liquid and in concentrated solutions, the hydroxyl group participates in the intermolecular hydrogen bond. It is characterized by a wide (3200-3600cm^{-1}) intense absorption maximum. Intramolecular bond is possible in hydroxyl-containing compounds. In dilute solutions, the formation of intermolecular hydrogen bonds is almost impossible. In the IR-spectrum of such solutions, the hydroxyl group is manifested by narrow intensive band, but for the bound hydroxyl group this band is located in the range of 3610-3620cm^{-1}, whereas for the HO-group forming an intramolecular bond, in the range of much lower frequencies ≈ 3500cm^{-1}.

The oscillation of unbound hydroxyl ν_{OH} in alcohols has a frequency of 3600-3615cm^{-1}, inherent in the hydrogen bond. Thus, the vibrational spectra can not only reliably prove the presence of a hydroxyl group, but also to determine its participation in the hydrogen bond. This allows you to solve a number of structural and analytical problems. The valence vibrations of the carbonyl group, regardless of which functional group it belongs to, are found to have an intense maximum in the range of 1650-1859 cm^{-1}, in which other bands are practically absent. These fluctuations are the most characteristic and allow us to confidently state the presence or absence of a carbonyl group in the substance. For carboxylic acids, the value of ν_{CO} is in the range of 1750-1770 cm^{-1}. Due to the induction effect (-1) decreases the bond length C=O, respectively, increases its power constant and wave number. The conjugation effect due to delocalization of π-electrons leads to a decrease in the wave number ν_{CO}. For systems C=C and C=O oscillations are in the range of wave numbers 1665-1685cm^{-1}. It means, that the nature of the corresponding functional group can be predicted from the wave number of the carbonyl group.

The experimental data agree well with the results of quantum chemical calculations of each SAS component separately and with the attribution of absorption bands of IR-spectrum to the corresponding oscillations of groups of atoms in molecules using the Hiperchem software package.

Conclusions

The stability and steadiness of the created systems largely depends on the correct selection of components, their concentrations and selected carrier. First of all, this applies to surfactants, their chemical structure and structure in solutions of different polarity and scientifically sound compositional selection of the composition, which is much more effective than individual substances. Glass beads are used as a carrier, which is chemically inert and mechanically strong, and the system of which it is part, stable to hydrocarbon liquids and completely impermeable to liquids and gases, and has a low thermal conductivity, resistance to cracking, satisfactory adhesion, low cost. The density of the developed protective coating does not exceed the density of the oil product, i.e. is lower than 0.6g/cm3. This condition is mandatory for the application of a floating coating on the surface of the petroleum product. The composition of the aerated hydrophilic coating, including polymer, nonionic surfactant, crosslinker and water, maximizes the electrical attraction between mixed surfactant micelles and polymer, without causing deposition of the coating, and the inclusion of gas-filled microspheres capable of providing excellent buoyancy of ultralight technological systems and their mechanical properties and also increasing the reliability of the protective barrier against the diffusion of hydrocarbon vapors during storage, high mechanical strength and fire safety. Thus, knowledge of physicochemical properties of components and features of interaction between them is essential not only for the correct combination of the best properties of components, but also for creation of composite systems with essentially new properties. A number of ultralight surfactant systems have been developed that interact with the surface of microspheres and prevent the evaporation of hydrocarbon liquids by 98-99.6%. both due to the chemical interaction of monooleatsorbitan polyethylene glycol with 2-(N-ethylper-fluorooctanesulfamide) acetate, and due to the formation of multicenter hydrogen bonds of this system with polyvinyl alcohol and the surface of the microspheres.

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Вплив фізико-хімічних параметрів компонентів поверхнево-активних систем на мінімізування випаровування вуглеводневих рідин

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Були розроблені високоефективні стабільні аеровані гідрофільні композиції, що містять фтортензиди та надлегкі мікродисперсні системи з використанням газонаповнених скляних, алюмосилікатних та полімерних мікросфер. Розробка композицій поверхнево-активних систем грунтується на поверхневій активності ПАР, їх розчинності у воді, здатності зв'язувати воду та утворення водневих зв'язків між компонентами. Основна умова стійкості покриття при змішуванні компонентів - хімічна взаємодія між ними та утворення системи, яка не розчиняється у вуглеводнях і не руйнується з точки зору використання. Найкращі плівкоутворюючі характеристики, необхідні для функціонування покриття (одночасне зменшення поверхневого натягу та утворення плівки) характерні для фторованих поверхнево-активних речовин (ФПАР), неполярна частина молекул яких містить фторвуглецевий ланцюг, завдяки чому вони нерозчинні у вуглеводнях, добре розчинні у воді і легко розподіляються по поверхні вуглеводневих рідин, створюючи захисну плівку. Вибір спів-ПАР ґрунтувався на здатності стабілізувати гідрофільні плівки на поверхні вуглеводнів не тільки при сприятливому гідрофільно-ліпофільному балансі, але і при найнижчих, хоча і вищих за критичну концентрацію міцелоутворення (ККМ), концентраціях для утворення змішаних адсорбційного шару підвищеної міцності. Цією поверхнево-активною речовиною був розчинний у воді і маслі твін-80, що значно збільшує гідрофільну частину та підвищує стабільність ПАС та стабільність аеросистеми. Введення скляних мікросфер у компоненти системи значно підвищило її міцність та стабільність.

При дослідженні стабільності та газопроникності розроблених систем поверхнево-активної речовини було встановлено, що ізоляційна здатність покриття зростає зі збільшенням гідрофільно-ліпофільного балансу системи та внаслідок хімічної взаємодії між карбоксильною групою фтортензиду та гідроксильними групами ПАР.

Ключові слова: випаровування вуглеводневих рідин, стабільність поверхнево-активної системи, гідрофільно-ліпофільний баланс, скляні сферичні мікрокульки