

Effect of Oligomers and Polymers of Ethylene Glycols on Micellar Characteristics of Aqueous Solutions of Sodium Pentadecyl Sulfonate

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Abstract: The influence of oligomers and polymers of ethylene glycols with a different molecular mass on the structural transformations of aqueous solutions of surfactant, sodium pentadecylsulfonate, studied by viscometry and light scattering methods depending on their content in the system. It established that ethylene glycol with a molecular mass of 2,000 and 40,000 does not affect the structure of the system. For ethylene glycol with a molecular mass of 4,000, 6,000, and 20,000, a clearly expressed complex course of the dependence of the intrinsic viscosity of the micelle system on the polymer content was established. It assumed that with the changes in the concentration of ethylene glycols in the system, micelles are compacted due to a change in the balance of hydrophobic-hydrophilic interactions. In parallel with the change in apparent micelle masses and asymmetry coefficients determined by the light scattering method, the intrinsic viscosity also changes depending on the system's composition. The practical application of this research is that it allows the performance properties of sodium pentadecyl sulfonate (SPDS) to be adjusted, thereby expanding and improving its applications.

Key-Words: - ethylene glycol (EG), polyethylene glycol (PEG), sodium pentadecylsulfonate (SPDS), viscometry, light scattering, surfactant.

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1 Introduction

This work is devoted to the study of the influence of various amounts of water-soluble oligomers and polymers of ethylene glycols with different molecular masses on the structural transformations of aqueous solutions of an anionic surfactant - sodium pentadecylsulfonate (SPDS). The effect of oligomers and polymers at various masses on the surfactants was studied by viscometry and light scattering methods.

Polyethylene glycol (PEG), also known as polyethylene oxide (PEO) or poly (oxyethylene) (POE) is a synthetic, hydrophilic, and biocompatible polyether. Typically, materials with molecular weights less than 20,000 g/mol are referred to as PEGs, whereas those with molecular weights above 20,000 g/mol are referred to as PEOs. These polymers are soluble in water as well as in many organic solvents, such as ethanol, acetonitrile, toluene, acetone, dichloromethane, hexane, and chloroform.

As is well known, the uses of surface-active substances (surfactants) have numerous industrial applications. Due to the property of self-

aggregation, they were used extensively in a variety of areas. They are used in detergents for cleaning processes, emulsions, waste-water treatment, lubricants for automobiles, electronic printing, biotechnology, cosmetics, petroleum industries, and more [1, 2, 3, 4]. Their performance can be bolstered with the addition of polymers. Surfactants and their micelles can change the surface tension, phase behavior, and rheology of a solution [2, 5, 9]. The addition of polymers particularly charged polymers has been shown to enhance bulk and interfacial properties for a variety of applications [6-10].

Currently, in various technological processes (in micelle-polymer flooding, emulsifying, foaming, and other solutions), polymers are used together with surfactants. Therefore, it is considered relevant to study the interaction of surfactants with macromolecular compounds to obtain information about the structural changes occurring in solutions and the possibility of their regulation. In addition, mixed surfactant-polymer systems are relatively simple analogs of biological structures that allow one to form model ideas about the mechanism of

self-assembly and functioning of bio membranes [11–13].

Many publications are devoted to the study of the interaction of polymers with low molecular mass anionic and cationic surfactants [14]. Most of them belong to polyelectrolyte (PE) complexes with oppositely charged surfactants. It is assumed that the volume of the PE macromolecule serves as the center of surfactant condensation, in which intramolecular micelle-like clusters formed. As the PE macromolecule is saturated with detergent ions, it is hydrophobized, compressed, and neutralized. When the critical charge density is reached, the macromolecule collapses.

Polyethylene glycol (PEG) is the simplest water-soluble non-ionic synthetic polymer, having a broad range of applications. It is used to cover the surfaces of colloidal particles to improve their biocompatibility since the modified surface shows increased resistance to the adsorption of protein [15, 16]. It is also used for covalent modification of biological macromolecules, peptides, liposomes, and other drug delivery systems [17–19]. Colloidal particles covered by PEG are not suppressed by the immune system, and their circulation time is increased up to 8–10 h, making them suitable for prolonged drug release [20 – 22]. Mixtures of PEG with anionic surfactants in aqueous solutions are used for various applications [23]. The solubility and colloidal properties of PEGylated particles depend strongly on the polymer molecular mass. Nanoparticles, oligomers, and low-molecular-weight compounds are usually covered with medium-sized polymers, with a mass of 20–50 kg/mol. Larger nanoparticles of sizes 50–100 nm are usually covered by low molecular weight PEG (3–10 kg/mol) since a further increase of the hydrodynamic radius leads to a reduction of the particle circulation lifetime [24, 25]. Small nanoparticles can be covered by low-molecular-weight PEG (1.5–20 kg/mol), leading to an increase in the lifetime of magnetic nanoparticles introduced into the body [26, 27]. PEG molecules having a size in the range of 2–8 kg/mol, at concentrations of 4–8 wt%, are used for the crystallization of proteins and biological macromolecules. In this process, the polymer occupies a large volume in the solution, displacing the protein. This induces the segregation and subsequent aggregation and formation of a solid crystalline phase [28].

2 Materials and Methods

We studied SPDS ($C_{15}H_{31}SO_3Na$) manufactured by VEB Leuna (Germany) and PEGN ($[-CH_2-CH_2-O-$

$]_n$) manufactured by "LOBA-Chemie" (Austria) without preliminary purification.

As known, PEG is a very common food supplement, used as a thickener in many fields. Surfactants are surface-active agents having the unique property to adsorb at the interface. This is due to the amphiphilic nature of surfactant monomers. They have a polar head group and a non-polar hydrophobic tail present in their structure. The surfactant monomers can aggregate to form colloidal-sized clusters in solutions, known as micelles. The formation of micelle occurs over a sharp range of concentrations of surfactant known as critical micelle concentration (CMC) due to the delicate balance between hydrophobic and hydrophilic interactions. The surfactant monomers dispersed in a solution below this concentration.

As is well known, viscometry is the leading method for determining average molecular mass in industrial applications. As a standalone method, it delivers values, typically by using an Ubbelohde capillary viscometer. In industrial applications, the measurement of $[\eta]$ provides a quick and easy route to the Molecules. For viscosity measurements, a minimum requirement is the knowledge of the dependence of $[\eta]$ on (M).

We carried out viscometric studies of solutions using a modified Ubbelohde viscometer with a hanging level, which allows the initial solution to be diluted directly in the viscometer itself; the capillary diameter was 0.56 mm. All measurements were carried out at temperature $295 \pm 0.01K$. The outflow time of the solvent-bidi distilled water under these conditions - was 101.1 sec. The initial concentration of the studied solutions was ~ 1.0 g/dl. Before measurements, both the solvent and the solutions were filtered with a Schott glass filter No.2. Intrinsic viscosity was found by double graphical extrapolation of values and infinite dilution.

The structure and interaction parameters of micelles in solutions of anionic surfactant PEG in water were studied by viscometry and light scattering methods. The effect of the addition of PEG, on micelle solutions, was investigated. A change in the power-law type behavior of the aggregation number on the SO concentration was observed at 2 wt. percentage of PEG in the surfactant/polymer mixtures. The screening effect concerning the micelle interaction took place with the addition of a large amount of PEG (about 10 wt. percentage in the mixture).

To study the above systems, light-dispersion studies were also used. Light scattering measurements were performed in the light scattering instrument FPS-3 (constructed by the Central

Constructional Bureau of Academy of Sciences, Russian Federation at an incident light length of $\lambda = 436$ nm.) with unpolarized incident 30–1500C with benzene as reference ($IU=48, 5 \times 10^{-6} \text{cm}^{-1}$). The refractive index increment ($\partial n/\partial C$) measured in a Pulfrich differential refractometer in bidistilled water by using a mercury-discharge lamp with blue light (435, 8 nm), giving $dn/dc = 0,109 \text{sm}^3\text{g}^{-1}$. To purify the solvent and solutions, they were successfully filtered through the glass filtered of different sizes (No 3-5) thermostated at 295K.

As is known, the light scattering of polymer mixture solutions has its characteristics [30-35]. Therefore, we limited ourselves to determining the apparent mass average micelle masses (\overline{M}_{app}), asymmetry coefficients ($[Z]$), and second virial coefficients (A_2) of the initial SPDS system and at the content of various amounts of PEG in the studied systems with the molecular mass of 40,000, 20,000 and 6,000. Table 1 shows the results of these studies.

Table 1.

Values \overline{M}_{app} , A_2 and $[Z]$ for PEG/surfactant mixtures of different compositions at different PEG molecular mass.

Mol. mass of PEG	$\frac{M_{PEG}}{M_{PDSN}}, \%$	$\overline{M}_{app} \times 10^{-5}$	$A_2 \times 10^4, \frac{\text{sm}^3}{\text{g}^2 \text{mol}}$	$[Z]$
40 000	100	0,28	1,20	1,62
	60	0,23	1,16	1,60
	40	0,19	1,07	1,41
	0	0,15	0,86	1,05
20 000	100	0,14	1,26	1,51
	60	0,21	1,24	1,44
	40	0,17	1,18	1,21
	0	0,15	0,86	1,05
6 000	100	0,07	1,24	1,21
	60	0,18	1,31	1,15
	40	0,16	1,20	1,08
	0	0,15	0,86	1,05

The mass average molecular mass (\overline{M}_{app}) was determined by both the methods of Zimm and Debye on the assumption about the shape of the macromolecules be a Gauss coil. Similar results were obtained. So, in a further investigation, the less complex Debye method was applied.

The refractive index increments ($\partial n/\partial C$) of the studied solutions were determined on a temperature-controlled Pulfrich IRF-23 refractometer at a temperature of $295 \pm 0.01 \text{K}$. The error in determining the refractive indices. The value was determined as the tangent of the slope of the linear dependence of the refractive index of the test solution on the concentration of the dissolved substance, expressed in g/cm^3 .

Light scattering studies were carried out on an FPS-3 photoelectric light scattering device designed by the Central Design Bureau of the Academy of Sciences of Russia at an incident light length of $\lambda = 436$ nm. The solvent used was bidistilled water at a temperature of $295 \pm 0.01 \text{K}$. Benzene was used as a calibration liquid, for the absolute value of the light scattering coefficient of which $J_u = 48.5 \cdot 10^{-6} \text{cm}^{-1}$ was taken (for incident natural light with a length of $\lambda = 436$ nm. In light-dispersion studies, solutions and solvents were purified by successive filtration through porous glass filters of various numbers (3–5) directly into a cuvette, which was pre-rinsed with the first portions of the filtrate. The samples studied by the light scattering method were transparent micellar solutions. It should note that the filtration of solutions, as well as work with them, required the development of a certain technique, since an aqueous solution of amphiphilic foams strongly during manipulations with the solution.

3 Results and Discussion

It is widely accepted [29, 30] that the viscosity of solutions is the measure of interaction between polymers and surfactants, and the formation of complexes between them. In this work, the influence of polyethylene glycols with a different molecular mass on the hydrodynamic behavior of aqueous solutions of SPDS is studied by the viscometry method.

Fig.1 shows the dependence of intrinsic viscosity ($[\eta]$) of SPDS-water one-micelle systems on the PEG content in the system. On the same graph, dotted lines are drawn between the values of the initial surfactant solution and pure PEG in water. It would correspond to the dependence on the composition of the mixture with non-interacting components of the mixture (it is assumed that the total viscosity of the mixture is the additive sum of the viscosity of the components).

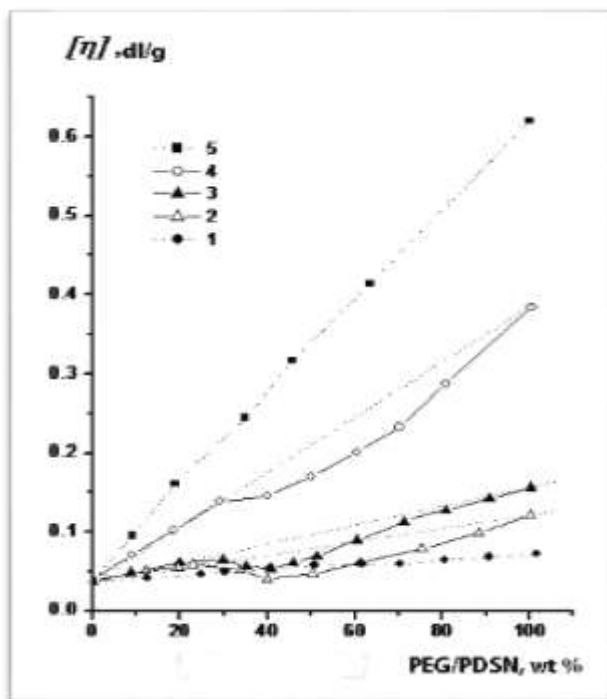


Fig.1: Dependence $[\eta]$ of micellar solutions of SPDS/water on the content in the PEG system with the following molecular masses: 1-2000, 2-4000, 3-6000, 4-20000, 5-40000.

As can be seen from the data presented (Fig.1), for PEGs with molecular masses of 2000 and 40000, linear dependences of intrinsic viscosities on the PEG/SPDS ratios are observed, indicating the absence of interactions between the components of the systems. Obviously, with an increase in the molecular weight of the polymer, after a certain value, due to the deterioration of the solubility of the polymer, macromolecules and free micelles coexist in the system, which does not interact with each other. At very low values of the molecular weight of the polymer, we do not exclude the interaction of macromolecules with micelles, but it is obvious that when small macromolecules aggregated on micelles, the shape and size at 40% content in the mixture of PEG. After this interval, growth was observed with increasing polymer concentration in the mixture. A negative deviation from the additive value, and even more so, the presence of a minimum in the dependence on the PEG content in the system indicates the presence of an interaction between the components of the systems.

Let us try to explain the observed picture. It is known that the interaction of a polymer with a surfactant reduces the hydration shell of the latter. In particular, in the works of Varshavsky et al., X-

ray diffraction proved that PEG has a pronounced water-binding property [36]. A decrease in the hydration shell of micelles entails the compaction of the latter and, accordingly, a negative deviation from the additive value.

Obviously, at lower PEG contents, the system contains free micelles and polymer coils. At certain polymer concentrations, only mixed aggregates exist in the system, polydisperse in terms of surfactant content. At high polymer concentrations, mixed aggregates and free polymer coils are present in the mixture. It can be assumed that with a change in the PEG concentration in the system, micelles are compacted due to a change in the balance of hydrophobic-hydrophilic interactions, because of which the above pattern of changes in the composition of the mixture is observed.

The same picture was established in a photoluminescence study of the interaction of sodium dodecyl sulfonate with water-soluble polymers (poly-N-vinylpyrrolidone, polyethylene oxide) [24]. It concluded that micelles of sodium dodecyl sulfonate bind to polymers, and the sizes of micelles become smaller than in the absence of the polymer [24].

The refractive index increments $(\partial n/\partial C)$ of the studied solutions were determined on a temperature-controlled Pulfrich IRF-23 refractometer at a temperature of $295 \pm 0.1K$. The error in determining the refractive indices $\pm 5 \times 10^{-5}$. The value was determined as the tangent of the slope of the linear dependence of the refractive index of the test solution on the concentration of the dissolved substance, expressed in g/cm^3 .

The solvent used was bidistilled water at a temperature $295 \pm 0.1K$. Benzene was used as a calibration liquid, for the absolute value of the light scattering coefficient of which $J_u = 48.5 \cdot 10^{-6} cm^{-1}$ was taken (for incident natural light with a length of $\lambda = 436 nm$). In light-dispersion studies, solutions and solvents purified by successive filtration through porous glass filters of various numbers (3-5) directly into a cuvette, which was pre-rinsed with the first portions of the filtrate. The samples studied by the light scattering method were transparent micellar solutions. It should be noted that the filtration of solutions, as well as work with them, required the development of a certain technique, since an aqueous solution of amphiphile foams strongly during manipulations with the solution.

Noteworthy is the pattern of change in $\overline{M}_{app} [Z]$, and A_2 , which in general show a symbiotic course

with the pattern of change [7] depending on the composition of the mixture. Comparison of the results of light-dispersion studies showed that, at a content of ~0.35% PEG in the system, the micelles of an aqueous solution of surfactant are maximally compacted.

4 Conclusion

Thus, the study of the effect of PEGs with different molecular masses on the structural transformations of aqueous solutions of PDSN depending on the polymer content in the system showed that PEGs with molecular masses of 2 000 and 40 000 have absolutely no effect on the structure of the system. For PEGs with molecular masses of 4 000, 6 000, and 20 000, a complex character of the dependence of the intrinsic viscosity of the micellar system on the polymer content in it was established. It assumed that with a change in the concentration of PEG in the system, micelles are compacted due to a change in the balance of hydrophilic-hydrophobic interactions. The practical application of this research is that it allows the performance properties of sodium pentadecyl sulfonate (SPDS) to be adjusted, thereby expanding and improving its applications. Naturally, with further research of this system, other additives will be tested to improve the performance properties of the studied polymer. This study is experimental. It is significantly more expensive than theoretical ones and usually allows you to obtain specific and reliable information in a fairly short time. In the future, similar studies are expected on other polymer objects to improve their performance properties. For almost 50 years, our laboratory has conducted similar research experiments and collected data from the results obtained. This method can be combined with artificial intelligence by processing this data and applying an appropriate machine learning model.

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