

STUDY OF THE SURFACE-BULK PROPERTIES OF AN ACRYLIC DISPERSION IN THE PRESENCE OF AN AMINE-CONTAINING ADDITIVE

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Abstract. *Introduction.* Waterborne coating materials based on polymer dispersions are regarded as a priority direction in coating technology due to reduced environmental impact and increasingly stringent requirements for production and application safety. However, aqueous acrylic dispersions retain technological limitations associated with film formation, interfacial interactions, and resistance to moisture exposure. One promising approach to improving performance is the incorporation of amine-containing additives capable of regulating charge–associative processes and the structure of the interfacial layer. The aim of this study was to determine the effect of an amine-containing additive on the surface–bulk properties of an aqueous acrylic dispersion and to identify the factors governing changes in interfacial activity and structural organization within the bulk phase. The research objectives included a comparative analysis of the “water–additive” system and the “acrylic dispersion–water–additive” system, as well as an examination of the mechanisms responsible for the redistribution of protonated and less ionized forms of the additive between the bulk phase and the phase boundary. *Methods.* Interfacial characteristics were evaluated using an ACAM series instrument equipped with video recording of the droplet profile and calculation of interfacial parameters by drop shape analysis based on the Young–Laplace equation. Bulk properties were monitored by conductometry (specific electrical conductivity) and potentiometry with pH measurements while varying the content of the amine-containing additive polyethylene polyamine. *Results and discussion.* It was shown that in an aqueous medium the additive exhibits limited interfacial activity. In the presence of an acrylic dispersion, the interfacial effect is enhanced, indicating the involvement of the additive in specific interactions with functional groups of the film-forming polymer and a change in its distribution between the surface and the bulk phase.

Conclusion. The results obtained substantiate the use of polyethylene polyamine as a tool for controlling interfacial and bulk processes in aqueous acrylic dispersions and may be applied in the development of waterborne coating systems.

Keywords: waterborne coating materials; acrylic dispersion; amine-containing additive; interfacial interactions; surface–bulk properties.

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Citation: Dyuryagina A.N., Ostrovnaya D.Yu., Byzova Yu.S., Lutsenko A.A. Study of the surface–bulk properties of an acrylic dispersion in the presence of an amine-containing additive. *Chem. J. Kaz.*, **2026**, 1(93), 69-78. DOI: <https://doi.org/10.51580/2026-1.2710-1185.07>

1. Introduction

In recent years, a steady global trend toward replacing solvent-borne coatings with water-dispersible systems has been observed [1]. This transition is primarily driven by increasingly stringent environmental regulations on volatile organic compound (VOC) emissions, corresponding changes in technical standards, and advances in polymer synthesis technologies that enable the design of materials with tailored properties [2].

According to Coherent Market Insights, in 2025 waterborne coatings accounted for approximately 40.5% of the total global paint and coatings production volume, reflecting a systematic shift in market demand toward environmentally safer systems with reduced emissions of hazardous substances [3]. Furthermore, estimates by Precedence Research indicate that the global market volume of waterborne coatings reached USD 96.15 billion in 2025, with a compound annual growth rate (CAGR) of 5.5% [3,4].

However, replacing organic solvents with water presents several technological challenges, as water exhibits fundamentally different physicochemical properties compared with organic solvents [5]. In addition, most conventional film-forming agents are insoluble in water, and only a limited number of water-soluble film formers are capable of producing transparent (true or molecular) solutions in aqueous media [6]. For this reason, aqueous dispersions of polymers and oligomers have become the most widely used systems, among which acrylic dispersions occupy a leading position [7].

Coatings based on acrylic film-forming agents are characterized by high weather resistance, including light, thermal, and chemical stability, as well as resistance to contamination and to acidic and alkaline environments [8,9]. At the same time, such coatings typically do not allow the production of high-solids formulations and may exhibit insufficient mechanical strength, limited water resistance, and relatively low corrosion resistance [10-12].

One of the main approaches to the targeted modification of the properties of polymer composite materials and their coatings is the use of additives exhibiting surfactant activity [13]. The selection of polyethylene polyamine as an additive in the present study was determined by two factors. First, the amphiphilic nature of its macromolecules, resulting from the presence of polar amine groups within a mixture of high-molecular-weight amines (characterized by the amine number) and hydrocarbon fragments, provides pronounced interfacial activity and the ability to participate in specific interactions within aqueous dispersion systems. Second, the additive ($M = 5000$) is water-soluble, which facilitates its incorporation into the aqueous phase [14-16].

From a physicochemical standpoint, modification induced by differences in the thermodynamic characteristics of the film-forming polymer and the additive is accompanied by concentration fluctuations of the introduced substance between the bulk phase of the aqueous polymer system and its interfacial surface energy at the air interface, primarily due to the development of adsorption processes. The variety of processes occurring in the bulk and interfacial phases differs only

slightly in terms of the energy required for their initiation [17]. The narrow energy range within which various states of the initial and modified forms are realized implies the necessity for precise and selective control of these processes in order to achieve the desired effects while preventing undesirable outcomes [18].

At the same time, understanding the physicochemical processes occurring in aqueous polymer dispersions is also important from a practical perspective. Changes in surface tension and electrokinetic characteristics of such systems can influence pigment wetting, dispersion stability, and film formation processes, which ultimately affect the performance properties of coatings, including adhesion, mechanical strength, and protective efficiency [19]. Therefore, the study of the influence of surface-active additives on the interfacial and bulk properties of aqueous polymer systems is relevant not only for fundamental research but also for the rational design of waterborne coating formulations.

In this context, it was considered appropriate to determine the effect of the additive concentration on the regularities of processes occurring both in the bulk of the aqueous acrylic dispersion (dissociation processes, hydrogen ion concentration, pH) and at its interface with air (surface tension).

2. Experimental Section

The objects of investigation were an aqueous acrylic dispersion based on the glossy acrylic varnish “Solax” and an amine-containing additive, polyethylene polyamine (PEPA), TU 2413-357-00203447-99. The studies were carried out using model compositions of the “water–PEPA” type and ternary systems of the “film former–water–PEPA” type, with the additive concentration varied within the range of 0–1.2 % in increments of 0.2 %. All solutions and compositions were prepared immediately prior to measurement and examined at a temperature of 295 K.

PEPA solutions of the required concentration were prepared individually: a weighed portion of the additive was measured on an analytical balance and dissolved in a specified volume of distilled water under stirring until complete homogenization. The ternary compositions were obtained by introducing PEPA into a calculated volume of the acrylic varnish “Solax” under stirring until a visually homogeneous state without observable aggregates was achieved. According to preliminary kinetic experiments, equilibrium in the investigated systems was established within 30 min; therefore, all subsequent measurements were performed after a standardized equilibration period of this duration.

The surface tension of the investigated systems was determined using an automated ACAM instrument by the pendant drop method (Figure 1). A droplet was formed at the tip of a dosing needle, its profile was recorded using a video system, and the surface tension was calculated by drop shape approximation in the instrument software based on the Young-Laplace equation. Measurements were carried out in parallel, and the results were averaged.



Figure 1 – Image of a water droplet dispensed for surface tension determination.

The specific electrical conductivity was measured using a “Multitest” conductometer. Prior to measurements, the instrument was calibrated with a standard solution. The measuring cell was rinsed with distilled water and with the test sample, after which conductivity was recorded once the readings stabilized at 295 K. The final value was taken as the average of three measurements.

The hydrogen ion concentration (pH) was determined using a “Multitest” potentiometer equipped with a combined glass electrode. Before measurements, calibration was performed using buffer solutions. The electrode was rinsed with distilled water and the test sample, and the pH value was recorded after stabilization of the readings at 295 K.

3. Results and Discussion

The results of the study are presented sequentially.

3.1. Surface Properties of Binary “Water–PEPA” Systems at the Air Interface

The surface tension isotherm ($T = 295$ K) of aqueous polyethylene polyamine solutions at the air interface demonstrates only minor changes in the liquid-gas surface tension (σ_{l-g}) (Figure 2). The maximum surface tension depression ($\Delta\sigma = 1.49$ mN/m) was observed at a PEPA concentration of 0.6 %. Beyond this concentration range ($C > 0.6$ %), the surface tension remains essentially constant ($\sigma = 71.28$ mN/m).

The weak surface activity of polyethylene polyamine compared with classical additives is due to the peculiarities of its chemical composition and structure, characterized by a large number of amine groups of different types ($-NH_2-$ and $-NH-$) and short hydrophobic fragments ($-CH_2-CH_2-$) [20,21]. The shift of the hydrophilic–lipophilic balance (HLB) toward polar groups reduces the tendency of PEPA to accumulate at the water–air interface. The quantitative dissociation characteristic of polyethylene polyamine, resulting from protonation of amine groups by water (accompanied by the formation of hydroxide ions), promotes hydrophilization and, consequently, the preferential localization of hydrated species within the bulk phase. The increase in the fraction of ionized

forms is clearly reflected in the observed changes in electrical conductivity (Figure 3a) and hydrogen ion concentration (Figure 3b) of aqueous polyethylene polyamine solutions.

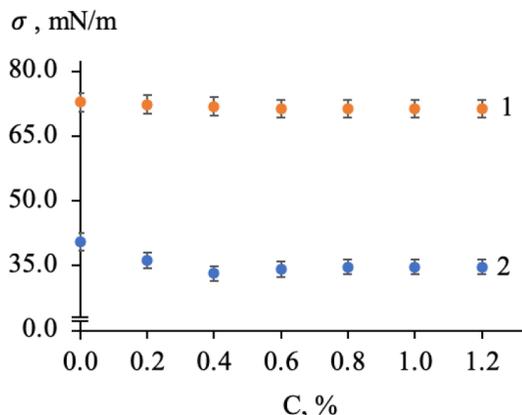


Figure 2 – Surface tension isotherms ($T = 295\text{ K}$) for the “water-PEPA” (1) and “film former-water-PEPA” (2) systems.

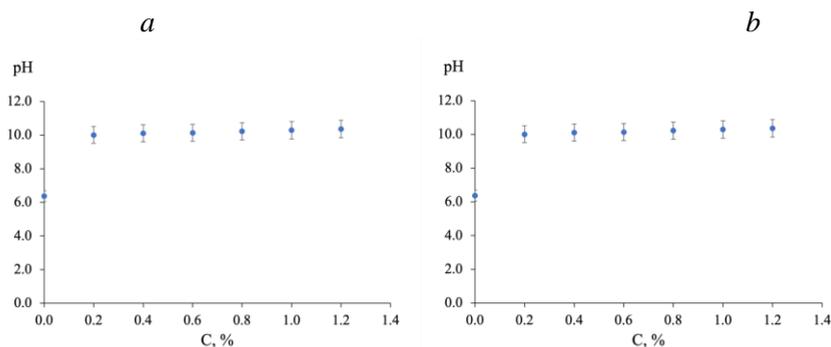


Figure 3 – Effect of polyethylene polyamine concentration in the binary “water-PEPA” system on: (a) specific electrical conductivity; (b) hydrogen ion concentration (pH).

As the polyethylene polyamine content increases up to 1.00–1.20 %, a continuous rise in solution conductivity (from 4.90 to 368.00 $\mu\text{S}/\text{cm}$) and pH (from 6.38 to 10.36) is observed. The pH shifts from near-neutral conditions ($\text{pH} = 6.38$) to an alkaline medium ($\text{pH} \geq 10$) as hydroxide groups accumulate in the aqueous phase, indicating a substantial increase in the fraction of ionized species.

3.2. Surface Properties of Ternary “Film Former–Water–Additive” Systems at the Air Interface

Upon introduction of PEPA into the film-forming solution, a different behavior is observed (Figure 4a and b).

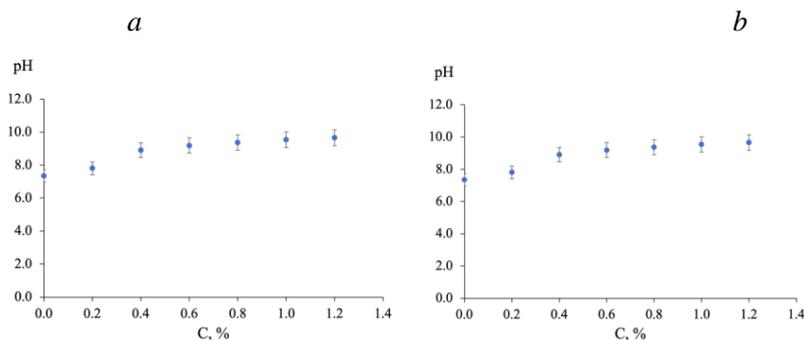


Figure 4 – Effect of polyethylene polyamine concentration in the ternary “film former-water-PEPA” system on: (a) specific electrical conductivity; (b) hydrogen ion concentration (pH).

In contrast to the behavior observed in the “water-PEPA” system, the increase in hydrogen ion concentration (pH) in the “film former-water-PEPA” system was accompanied by the opposite effect – a decrease in electrical conductivity as the additive concentration increased (Fig. 4a and b).

Moreover, the absolute values of conductivity (χ) in the ternary system were tens of times lower than those recorded for the binary system. This is confirmed by the relative conductivity values ω (1):

$$\omega = \chi_1 / \chi_2, \quad (1)$$

where χ_1 is the electrical conductivity of the binary “water-additive” system, and χ_2 is the electrical conductivity of the ternary “film former-water-additive” system.

Table 1 – Effect of additive concentration on the ratio of conductivity values ω

$C_{PEPA}, \%$	0.2	0.4	0.6	0.8	1.0	1.2
ω	30.54	43.63	56.76	64.52	74.05	81.06

The resulting effect of acrylic binder macromolecules in the “film former-water-PEPA” system, in which the water content is 69 %, on the surface activity of polyethylene polyamine is clearly illustrated by the surface tension isotherm (Fig. 1, curve 2). In the region of low additive concentrations ($C_{PEPA} \leq 0.4 \%$), the polymer present in the composition induces the migration of non-ionized forms of the polyelectrolyte to the air interface. This is evidenced by the observed decrease in specific surface energy. The maximum reduction in surface tension ($\Delta\sigma = 14.56 \text{ mN/m}$) was recorded at $C_{PEPA} = 0.4 \%$. This decrease in specific surface energy was observed in a weakly alkaline medium (pH = 8.91) at a specific electrical conductivity of 5.57 mS/cm (Fig. 3a and b).

Further addition of polyethylene polyamine (above 0.4 %) promoted its preferential localization in the bulk liquid phase rather than at the air interface:

instead of a continued decrease, the surface tension remained constant at 34.03 ± 0.07 mN/m (plateau on curve 2, Fig. 1). The appearance of a plateau at higher PEPA concentrations indicates the development of association processes, accompanied by a decrease in conductivity values (Fig. 3a) and an increase in pH (Fig. 3b).

Spontaneous association of the acrylic film-forming macromolecules and the polyelectrolyte upon reaching a critical association concentration (onset of the plateau on curve 2, Fig. 1) is likely driven by electrostatic interactions between ionized species, hydrogen bonding, and van der Waals forces. As the additive concentration increases further, the degree of association rises and the mobility of larger associates decreases, which accounts not only for the absence of surface activity ($\sigma_{l-g} = \text{const}$) but also for the observed reduction in specific electrical conductivity (Fig. 3a).

4. Conclusion

In the binary “water–PEPA” system, polyethylene polyamine exhibits weak interfacial activity due to its quantitative dissociation resulting from protonation of amine groups by water, accompanied by hydroxide ion formation. As the PEPA concentration increases to 1.00–1.20 %, a steady rise in conductivity from 4.90 to 368.00 $\mu\text{S}/\text{cm}$ and a simultaneous increase in pH from 6.38 to 10.36 are observed.

In the ternary “film former–water–PEPA” system, the reduced extent of PEPA dissociation due to the lower solvent content (69 %) enhances its surface activity at the air interface. The maximum reduction in surface tension ($\Delta\sigma = 14.56$ mN/m) was recorded at a PEPA concentration of 0.4 %. The highest surface activity effect was achieved in the weakly alkaline region (pH = 8.91) at a specific electrical conductivity of 5.57 mS/m.

Conflict of interest: The authors declare that there is no conflict of interest between the authors that requires disclosure in this article.

ҚҰРАМЫНДА АМИН БАР ҚОСПАНЫҢ ҚАТЫСУЫМЕН АКРИЛ ДИСПЕРСИЯСЫНЫҢ БЕТТІК-КӨЛЕМДІК ҚАСИЕТТЕРІН ЗЕРТТЕУ

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Түйіндемe. *Кіріспе.* Полимерлік дисперсиялар негізіндегі су арқылы сұйылтылатын лак-бояу материалдары экологиялық жүктемені төмендету және өндіру мен қолдану қауіпсіздігіне қойылатын талаптардың артуына байланысты жабындарды дамытудың басым бағыты ретінде қарастырылады. Сонымен қатар су негізіндегі акрил дисперсиялары плёнка түзілуі, фазалар аралық өзара әрекеттесулер және ылғалдың әсеріне төзімділікке байланысты технологиялық шектеулерін сақтайды. Қасиеттерді жақсартудың перспективалы бағыттарының бірі – зарядтық-ассоциациялық процестерді және фазалар аралық қабаттың құрылымын реттеуге қабілетті аминқұрамды беткі-белсенді заттарды енгізу. *Жұмыстың мақсаты* су негізіндегі акрил

дисперсиясының беткі және көлемдік қасиеттеріне аминқұрамды қоспаның әсерін анықтау және фазалар аралық белсенділік пен компоненттердің көлемде құрылымдануының өзгеруін айқындайтын факторларды белгілеу болды. Зерттеу міндеттеріне «су–қоспа» және «акрил дисперсиясы–су–қоспа» жүйелерінің мінез-құлқын салыстыру, сондай-ақ протондалған және аз нондалған қоспа формаларының көлем мен фазалар аралық шекара арасында қайта бөліну себептерін талдау кірді. *Әдістері.* Фазалар аралық сипаттамалар АСАМ сериялы аспаптарда тамшы профилін бейнетіркеу және Юнг-Лаплас теңдеуіне негізделген тамшы пішінін аппроксимациялау әдісі арқылы есептелді. Көлемдік қасиеттер кондуктометрия (меншікті электрөткізгіштік) және потенциометрия әдістерімен, аминқұрамды қоспа – полиэтиленполиаминнің мөлшерін өзгерте отырып, сутектік көрсеткішті (рН) өлшеу арқылы бақылауға алынды. *Нәтижелер мен талқылау.* Су ортасында қоспаның фазалар аралық белсенділігі шектеулі екені көрсетілді. Акрил дисперсиясының қатысуында фазалар аралық әсердің күшеюі байқалады, бұл қоспаның плёнка түзушінің функционалдық топтарымен спецификалық өзара әрекеттесулерге тартылуын және оның бет пен көлем арасында таралуының өзгеруін көрсетеді. *Қорытынды.* Алынған нәтижелер полиэтиленполиаминді су негізіндегі акрил дисперсияларындағы фазалар аралық және көлемдік процестерді басқарудың тиімді құралы ретінде қолдануды негіздейді және су арқылы сұйылтылатын жабындарды әзірлеуде пайдаланылуы мүмкін.

Түйін сөздер: су-дисперсиялы лак-бояу материалдары; акрил дисперсиясы; аминқұрамды қоспа; фазалар аралық өзара әрекеттесулер; беткі-көлемдік қасиеттер.

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ИССЛЕДОВАНИЕ ПОВЕРХНОСТНО-ОБЪЕМНЫХ СВОЙСТВ АКРИЛОВОЙ ДИСПЕРСИИ В ПРИСУТСТВИИ АМИНОСОДЕРЖАЩЕГО АДДИТИВА

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Резюме. *Введение.* Водоразбавляемые лакокрасочные материалы на основе полимерных дисперсий рассматриваются как приоритетное направление развития покрытий вследствие снижения экологической нагрузки и повышения требований к безопасности производства и применения. Вместе с тем водные акриловые дисперсии сохраняют технологические ограничения, связанные с формированием плёнки, межфазными взаимодействиями и устойчивостью к воздействию влаги. Одним из перспективных путей улучшения свойств является введение аминоксодержащих поверхностно-активных веществ, способных регулировать зарядово-ассоциативные процессы и структуру межфазного слоя. *Целью работы* являлось установление влияния аминоксодержащей добавки на поверхностно-объёмные свойства водной акриловой дисперсии и выявление факторов, определяющих изменение межфазной активности и структурирования компонентов в объёме. *Задачи исследования* включали сопоставление поведения системы «вода–аддитив» и системы «акриловая дисперсия–вода– аддитив», а также анализ причин перераспределения протонизированных и менее ионизированных форм добавки между объёмом и границей раздела фаз. *Методы:* межфазные характеристики оценивали на установке серии приборов АСАМ с видеорегистрацией профиля капли и расчётом параметров межфазного состояния методом аппроксимации формы капли на основе уравнения Юнга-Лапласа. Объёмные свойства контролировали методами кондуктометрии (удельная электропроводность) и потенциометрии с измерением водородного показателя (рН) при варьировании содержания аминоксодержащей добавки полиэтиленполиамин. *Результаты и обсуждение:* показано, что в водной среде добавка

проявляет ограниченную межфазную активность. В присутствии акриловой дисперсии межфазный эффект усиливается, что указывает на вовлечение добавки в специфические взаимодействия с функциональными группами плёнкообразующего и на изменение её распределения между поверхностью и объёмом. *Заключение.* Полученные результаты обосновывают использование полиэтиленполиамина как инструмента управления межфазными и объёмными процессами в водных акриловых дисперсиях и могут быть применены при разработке водоразбавляемых покрытий.

Ключевые слова: водно-дисперсионные лакокрасочные материалы; акриловая дисперсия; аминоксодержащий аддитив; межфазные взаимодействия, поверхностно-объёмные свойства.

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