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THE INFLUENCE OF Ph OF THE ENVIRONMENT ON SOIL STABILITY

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Abstract. In world practice, the process of structure formation of mineral dispersions with polymers is one of the effective methods for solving many environmental problems. Patent research over the past 15 years has shown that the developments available in this area relate mainly to the structure formation of soils and soils with salts of low molecular weight substances and polymers. The main disadvantages of modern structure formers are the high salt content in the polycomplex formulation, insufficient water resistance, low aggregation ability and unsatisfactory adhesive properties. Soil particles having a diameter of 12 microns, in the pH range 10.0 - 3.0, the soil suspension should be practically stable, since the value of the energy barrier significantly exceeds the energy of thermal movement of the particles and is 166 kT (pH 10.0), 125 kT (pH 8.0), 63 kT (pH 6.0) and 41kT (pH 5.0). A further decrease in pH leads to a natural decrease in the barrier, which at pH 4.0 is 6 kT, and completely disappears at pH 3.0. Thus, for a fraction of a soil suspension with a diameter less than 12 µm, noticeable coagulation can be observed at pH £ 4.0. It is easy to show that larger soil particles with a diameter of 30-33 microns should be practically aggregatively stable even at pH 3.0, since the energy barrier in this case is 10-15 kT. When interacting with the smallest particles (a = 1 µm), the energy barrier values are: 44 kT (pH 10.0), 28 kT (pH 8.0), 18 kT (pH 6.0), 7 kT (pH 5.0) and 2 kT (pH 4.0) and only for these particles at pH < 4.0 can one expect a noticeable loss of aggregation stability by the dispersion. However, the experiment shows that significant coagulation of particles occurs already at pH 5.0 and continuously increases as the pH decreases.

Key words: structure-forming agent; soil; flocculation; coagulation; interaction energy; optical density; potentiometric titration; electrophoretic mobility.

Introduction. The formation of structure in clay (soil) suspensions is closely related to the processes of coagulation and flocculation [1-5]. Coagulation adhesion of particles occurs through the so-called short-range interaction (in the first potential minimum) along the most lyophobic areas of the surface, which are least protected by the solvation shells of the medium, and, as a result of further interaction of two particles, fixed at a distance of about 100 nm and which corresponds to the second minimum at the potential crooked. In this case, a kind of flexible connection is established - the so-called «pair» of particles, which cannot help but come close, not separate, and perform joint Brownian motion. Moreover, the fixation of particles in the second minimum with a sufficient concentration of the dispersed phase leads to the transition of the system from a state in the form of a sol to a structured system.

The presence of bound water in places of coagulation adhesion [6-12] prevents further convergence of particles, which causes a small adhesion force (speaking of adhesion force, we mean contact strength – one of the most important characteristics of dispersed structures, which determines almost all of its mechanical properties).

Soil structuring is possible with both low molecular weight compounds and high molecular weight reagents. Numerous studies show the advantage of using high-molecular substances: surfactants, polymers, polyelectrolytes, compositions of high-molecular compounds with salts of low-molecular compounds. In this regard, there is a new direction of colloidal chemical science, based on the concept of soil as a complex micro heterogeneous disperse system, which makes it possible to apply the principles of physicochemical mechanics of disperse systems using polyelectrolytes and surfactants to the processes of structure formation in soil dispersions.

Surfactants are organic compounds, the molecule of which contains both a polar group and a non-polar hydrocarbon chain, which has the property of surface activity. The presence of ionizing groups in the polymer units determines its properties as a polyelectrolyte [13].

As is known, electrostatic forces act between the links of a polyelectrolyte chain, the potential of which is determined by the degree of ionization [14-17]. If the degree of ionization is low, then the chain of the polymer/polyelectrolyte macromolecule statically twists due to internal rotations. Ionization of the chain leads to the appearance of repulsive forces equivalent to the external tensile force. As a result of this, macromolecules stretch, acquiring a helical transform. Thus, internal rotation, exhibiting micro-Brownian thermal motion within the polymer chain, pH changes can lead to all sorts of configurations. The state of the macromolecular chain that best corresponds to the maximum entropy corresponds to its twisting into a ball. The unfolding of the chain helps to reduce entropy, resulting in an elastic rotating force. Certain physicochemical conditions and the variety of rotational isomeric configurations of high molecular weight compounds provide additional opportunities for conformational changes. The presence of some parts of isomers in polar groups of atoms capable of forming a hydrogen bond causes stabilization by intermolecular interactions and leads to a fixed change in the configuration of the molecule.

Materials and research methods. The soil of the Azgir test site was used as the object of study. The soil cover of this territory is represented by brown and alkaline soils. The profile of brown soils is characterized by a clear differentiation into genetic horizons: humus, carbonate and salt. Due to the formation of complexes with solonetzes, salt-marsh and claypans, brown soils are characterized by the absence of humus in their composition and strong salinity. The thickness of the soil layer in the described area is very insignificant (2-10 cm).

The optical density of solutions was determined on an SF-46 spectrophotometer with an accuracy of ±2%. All experiments were carried out in cuvettes with an absorbing layer thickness of 1 cm.

Potentiometric titration was carried out in a thermostated cell using an I-500 ion meter with glass and silver chloride electrodes with an accuracy of ±0.2. The titration was carried out with constant stirring.

The electrophoretic mobility of solutions of the complex was measured in a modified Rabinovich and Fodiman device using the moving boundary method. The modification is that the cell is devoid of any taps, which allows measurements to be taken under thermostatic conditions. A potassium chloride solution with electrical conductivity equal to the electrical conductivity of the solutions under study was used as a side liquid.

Results and Discussion. Issues of the structure and physicochemistry of soil, which occupy one of the leading places among natural materials, are of great general scientific and applied importance. The possibility of more rational use of it in various branches of agriculture depends on their solution. However, the study of electrical surface properties and, especially, the aggregative stability of soil dispersions have so far received insufficient attention, whereas they often determine many other properties of soil dispersions.

To study the effect of pH on the electro surface properties and aggregative stability of an aqueous soil suspension, soil particles with a diameter of less than 70 μm were taken. The study of the aggregative stability of the soil dispersion was carried out as follows: 0.5 g of soil was placed in 100 ml of distilled water and then, by dilution, a working solution was prepared with a concentration of 0.1% and a given pH value, which was regulated by introducing an acid or alkali.

The effect of pH on the aggregative stability of soil dispersion was characterized by the values $(\Delta D/ D_o)\cdot 100$ %, where $\Delta D = D_o - D_\tau$, D_o, and D_τ are the optical densities of the dispersion at time t in water (pH 8) and in the presence of acid and alkali (pH 10.0, 6.0–3.0), respectively. The electrophoretic mobility of soil particles (fractions smaller than 12 µm in diameter) was determined by macro electrophoresis. The electro kinetic potential was calculated using the Helmholtz-Smoluchowski formula.

The dependence of the interaction energy of soil particles on the distance between them was calculated as follows. In the case of particles of equal diameter, the ion-electrostatic component was calculated using formula (1):

$$U_{\acute{Y}} = \frac{\varepsilon \cdot \cdot \grave{a} \cdot \psi^2}{4} \cdot \left\{ In\left[\frac{1+\exp(-\kappa \cdot H)}{1-\exp(-\kappa \cdot H)}\right] + In\left[1-\exp(-2\kappa H)\right] \right\} \tag{1}$$

In the case of particles differing in size (a_1 and a_2), formula (2) was used:

$$U_{\acute{Y}} = \frac{\varepsilon \cdot \cdot \grave{a}_1 \cdot \grave{a}_2 \cdot \psi^2}{2(\grave{a}_1 + \grave{a}_2)} \cdot \left\{ In\left[\frac{1+\exp(-\kappa \cdot H)}{1-\exp(-\kappa \cdot H)}\right] + In\left[1-\exp(-2\kappa H)\right] \right\} \tag{2}$$

ε - the dielectric constant of the dispersion medium; a – the radius of the particle; ψ – Stern potential; H – distance between particles; κ – Debye parameter: $\kappa = \sqrt{\dfrac{2n \cdot z^2 \cdot e^2}{\varepsilon \cdot \varepsilon_0}}$, where n – electrolyte concentration in solution; ε_o – electrical constant; z – the charge of the electrolyte counterion; e – electron charge. In the calculations it was assumed that the y potentials of the particles are equal to their electrokinetic potentials.

The calculation of the molecular component of the interaction energy of particles of equal size was carried out according to microscopic theory using formula (3):

$$U_M = -\frac{A_{121} \cdot a \cdot \lambda}{12 \cdot H \cdot \lambda_1 + 3{,}54 \cdot \pi \cdot H} \quad \text{in } H < 15 \text{ нм} \tag{3}$$

$$U_M = -\frac{A_{121} \cdot a}{\pi}\left(\frac{2{,}45 \cdot \lambda}{120 \cdot H^2} + \frac{\lambda^2}{1045 \cdot H^3} + \frac{\lambda^3}{5{,}62 \cdot 10^4 \cdot H^4}\right) \quad \text{in } H > 15 \text{ нм} \tag{4}$$

For particles differing in size, formula (2) was used:

$$U_M = -\frac{A \cdot a_1 \cdot \grave{a}_2}{6 \cdot H \cdot (\grave{a}_1 + \grave{a}_2)(1 + 1{,}77p)} \quad \text{in } 0 < p < 2 \tag{5}$$

$$U_M = -\frac{2A \cdot a_1 \cdot \grave{a}_2}{H \cdot (\grave{a}_1 + \grave{a}_2)}\left(\frac{2{,}45}{60p} + \frac{2{,}17}{180p^2} - \frac{0{,}59}{420p^3}\right) \quad \text{in } 0{,}5 < p < \infty \tag{6}$$

where $p = 2\pi \cdot H/\lambda$, A_{121} – the Hamaker constant for soil particles in water; λ – the characteristic wavelength (λ = 100 nm). The Hamaker constant $A_{121} = 1{,}6 \cdot 10^{-13}$ erg was calculated using the formula:

$$A_{121} = (\sqrt{A_{11}} - \sqrt{A_{22}})^2 \tag{7}$$

where A_{11} – the Hamaker constant for soil particles (accepted according to (4) equal to 1.2×10^{-13} erg); A_{22} – the Hamaker constant for water (accepted according to (3) equal to 4.38×10^{-13} erg).

The obtained values of the electro kinetic potential depending on the pH of the medium are presented in Fig. 1. As follows from Fig. 1, throughout the entire pH range ζ - potential of soil particles in water is negative. The isoelectric point of soil particles is at pH 2.0 (Fig. 1).

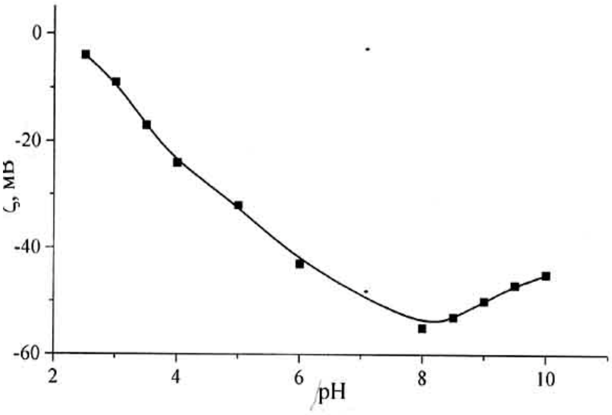
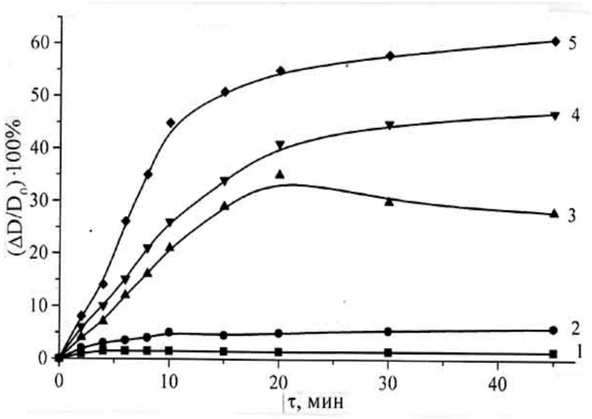


Figure 1 – Dependence of the electro kinetic potential of soil particles on the pH of the solution

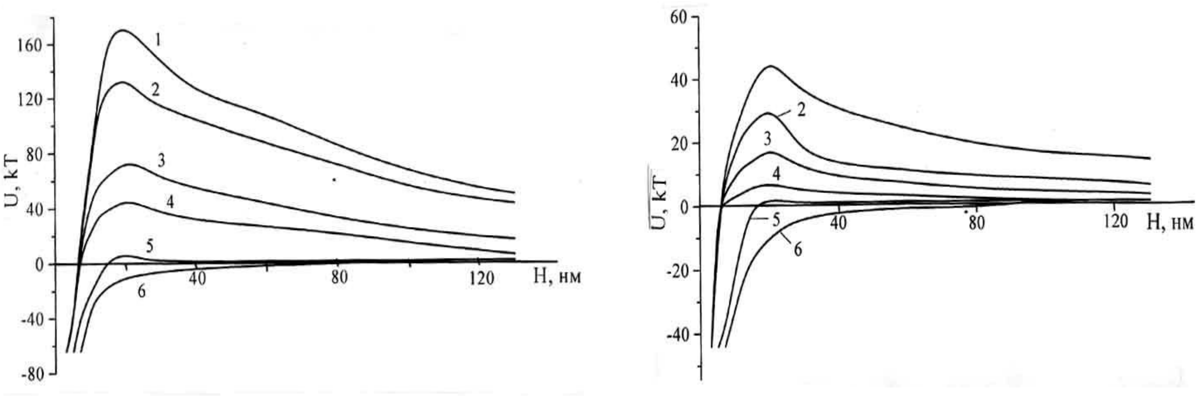
Dependences of ΔD/ D₀ on time τ in the pH range 3.0 – 10.0 are presented in Fig. 2. At pH 8.0 and 10.0, the soil suspension is aggregatively stable (low values of ΔD/ D₀) throughout the studied time interval.



1 – 8,0; 2 – 10,0; 3 – 6,0; 4 – 4,0; 5 –3,0

Figure 2 – Dependence of ΔD/D₀ on time for a soil suspension (diameter less than 12 μm) at different pH values

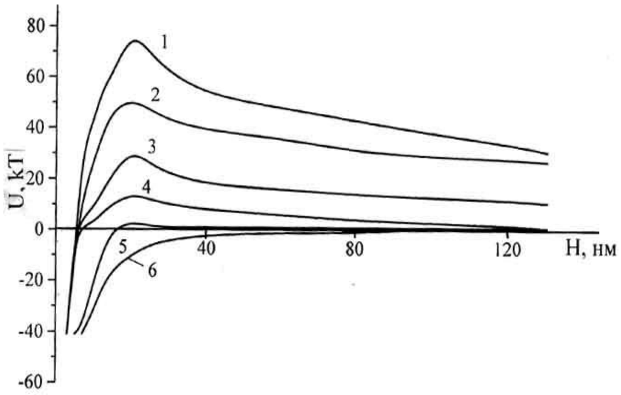
A further decrease in pH leads to an increase in ΔD/ D₀, which indicates the aggregation of soil particles and an increase in the rate of their sedimentation. This effect naturally increases as the pH decreases. To explain the results obtained on the effect of pH on the aggregative stability of the soil suspension, calculations were carried out of the interaction energy of identical (2 μm - 2 μm and 12 μm - 12 μm) and different (33 μm - 2 μm) particle diameters as a function of different distances between them (Fig. 3, 4).



a
1 – 10,0; 2 – 8,0; 3 – 6,0; 4 – 5,0; 5 – 4,0; 6 – 3,0

b
1 – 10,0; 2 – 8,0; 3 – 6,0; 4 – 5,0; 5 – 4,0; 6 – 3,0

Figure 3 – Dependence of the interaction energy of soil suspension particles of equal diameter on distance at different pH values. The average particle diameter is 12 µm (a) and 2 µm (b)

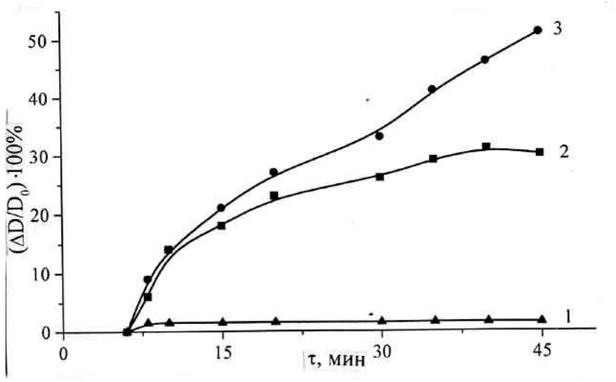


1 – 10,0; 2 – 8,0; 3 – 6,0; 4 – 5,0; 5 – 4,0; 6 – 3,0

Figure 4 – Dependence of the interaction energy of soil suspension particles of different diameters (33 µm – 2 µm) on distance at different pH values of the environment

From the results it follows that in the case of particles having a diameter of 12 microns, in the pH range 10.0 – 3.0, the soil suspension should be practically stable, since the value of the energy barrier significantly exceeds the energy of thermal movement of the particles and is 166 kT (pH 10, 0), 125 kT (pH 8.0), 63 kT (pH 6.0) and 41 kT (pH 5.0). A further decrease in pH leads to a natural decrease in the barrier, which at pH 4.0 is 6 kT, and completely disappears at pH 3.0. Thus, for a fraction of a soil suspension with a diameter less than 12 µm, noticeable coagulation can be observed at pH ≤ 4.0. It is easy to show that larger soil particles with a diameter of 30-33 µm should be practically aggregatively stable even at pH 3.0, since the energy barrier in this case is 10-15 kT. When interacting with the smallest particles (a = 1 µm), the energy barrier values are: 44 kT (pH 10.0), 28 kT (pH 8.0), 18 kT (pH 6.0), 7 kT (pH 5.0) and 2 kT (pH 4.0) and only for these particles at pH < 4.0 can one expect a noticeable loss of aggregation stability by the dispersion. The experiment, however, shows (Fig. 3, a) that significant coagulation of particles occurs already at pH 5.0 and continuously increases as the pH decreases.

The emerging contradiction between the experiment and the calculation results can be eliminated if we attract ideas about the possibility of the process of orthokinetic coagulation occurring during the aggregation of rapidly settling large particles with smaller ones. As an example, Figure 4 shows the dependences of the interaction energy of soil particles of different diameters (33 µm and 2 µm) on the distance between them. In this case, the value of the energy barrier, determined by the size of the smaller particle, is significantly lower than in the case of the interaction of two particles with a diameter of 2 µm, and at pH 10.0, 8.0, 6.0, 5.0 and 4.0 are 75, 48, 31, 13 and 2 kT respectively. The calculation of the kinetic energy of settling soil particles with a diameter of 33 µm shows that it is ~ 50 kT and exceeds the energy barrier that exists during the interaction of particles with diameters of 33 and 2 µm at pH ≤ 5.0. Thus, in this pH range there is a fundamental possibility of overcoming it and undergoing the process of orthokinetic coagulation. The sharp drop in the kinetic energy of particles with a decrease in their size ($V_{\text{кин.}} \sim a^7$) suggests that in the case of soil particles with a diameter of less than 50 µm, the effect of orthokinetic coagulation should be insignificant. To confirm the last assumption, particles with a diameter of more than 20 µm were removed from the studied soil dispersion by settling. The results obtained on the effect of pH on the aggregative stability of soil dispersion less than 20 µm are presented in Fig. 5.



1 – 10,0, 8,0, 6,0; 2 – 4,0; 3 – 3,0

Figure 5 – Dependence of ΔD/D₀ on time for a soil suspension (diameter less than 25 µm) at different pH values

As follows from Fig. 5, the system is stable at pH 10.0 - 6.0 and begins to coagulate at pH 3.0, which is in good agreement with calculations of particle interaction energy performed on the basis of the DLVO theory.

From the above experiments it follows that the stability of aqueous soil dispersions with particle diameters less than 60 µm significantly depends on the pH of the medium and their behavior can be explained on the basis of the DLVO theory using the concepts of perikinetic and orthokinetic coagulation.

Conclusion. From the results it follows that in the case of particles with a diameter of 12 microns, in the pH range 10.0 - 3.0, the soil suspension should be practically stable, since the value of the energy barrier significantly exceeds the energy of thermal motion of the particles. Thus, for a fraction of a soil suspension with a diameter less than 12 µm, noticeable coagulation can be observed at pH ≤ 4.0.

The emerging contradiction between the experiment and the calculation results can be eliminated if we attract ideas about the possibility of the process of orthokinetic coagulation occurring during the aggregation of rapidly settling large particles with smaller ones. interaction energy of soil particles differing in diameter (33 µm and 2 µm) from the distance between them, the value of the energy barrier, determined by the size of the smaller particle, is significantly lower than in the case of interaction of two particles with a diameter of 2 µm. The calculation of the kinetic energy of settling soil particles with a diameter of 33 microns exceeds the energy barrier that exists during the interaction of particles with diameters of 33 and 2 microns at pH ≤ 5.0. Thus, in this pH range there is a fundamental possibility of overcoming it and undergoing the process of orthokinetic coagulation. The sharp drop in the kinetic energy of particles with a decrease in their size suggests that in the case of soil particles having a diameter of less than 50 μm, the effect of orthokinetic coagulation should be insignificant.

The stability of aqueous soil dispersions with particle diameters less than 60 μm significantly depends on the pH of the environment and their behavior can be explained on the basis of the DLVO theory using the concepts of perikinetic and orthokinetic coagulation.

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ТОПЫРАҚ ТҰРАҚТЫҒЫНА pH ОРТАСЫНЫҢ ӘСЕРІ

Аңдатпа. Әлемдік тәжірибеде полимерлермен минералды дисперсиялардың құрылымын қалыптастыру процесі көптеген экологиялық мәселелерді шешудің тиімді әдістерінің бірі болып табылады. Соңғы 15 жылдағы патенттік зерттеулер көрсеткендей, осы саладағы әзірлемелер негізінен төмен молекулалық заттар мен полимерлердің тұздары бар топырақтар мен топырақтардың құрылымын қалыптастыруға қатысты. Қазіргі заманғы құрылымды қалыптастырушылардың негізгі кемшіліктері поликомплексті құрамдағы тұздың жоғары болуы, судың жеткіліксіз тұрақтылығы, төмен агрегация қабілеті және қанағаттанарлықсыз жабысқақ қасиеттері болып табылады. Диаметрі 12 мкм болатын топырақ бөлшектері, pH 10,0 – 3,0 диапазонында топырақ суспензиясы іс жүзінде тұрақты болуы керек, өйткені энергетикалық кедергінің мәні бөлшектердің жылулық қозғалысының энергиясынан айтарлықтай асып түседі және 166 кТ (pH 10,0), 125 кТ (pH 8,0), 63 кТ (pH 6,0) және 41 кТ (pH 5,0) құрайды. pH-ның одан әрі төмендеуі тосқауылдың табиғи төмендеуіне әкеледі, ол pH 4,0 кезінде 6 кТ, ал pH 3,0 кезінде толығымен жойылады. Осылайша, диаметрі 12 мкм-ден аз топырақ суспензиясының бір бөлігі үшін pH ≤ 4,0 кезінде айтарлықтай коагуляцияны байқауға болады. Диаметрі 30-33 мкм болатын үлкенірек топырақ бөлшектері pH 3,0 кезінде де іс жүзінде агрегаттық тұрақты болуы керек екенін көрсету оңай, өйткені бұл жағдайда энергетикалық кедергі 10-15 кТ. Ең кішкентай бөлшектермен (a = 1 мкм) әрекеттесу кезінде энергетикалық кедергі мәндері: 44 кТ (pH 10,0), 28 кТ (pH 8,0), 18 кТ (pH 6,0), 7 кТ (pH 5,0) және 2 кТ (pH 4,0) және тек осы бөлшектер үшін pH < 4,0 кезінде дисперсия арқылы агрегация тұрақтылығының айтарлықтай жоғалуын күтуге болады. Дегенмен, эксперимент бөлшектердің айтарлықтай коагуляциясы pH 5,0 кезінде орын алатынын және pH төмендеген сайын үздіксіз арта түсетінін көрсетеді.

Кілт сөздер: құрылым түзуші; топырақ; флокуляция; коагуляция; өзара әрекеттесу энергиясы; оптикалық тығыздық; потенциометриялық титрлеу; электрофорездік қозғалғыштық.

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ВЛИЯНИЕ pH СРЕДЫ НА УСТОЙЧИВОСТЬ ПОЧВЫ

Аннотация. В мировой практике процесс структурообразования минеральных дисперсии полимерами является одним из эффективных методов решения многих экологических проблем. Патентные исследования последних 15 лет показали, что имеющиеся в данной области разработки касаются, в основном, структурообразования почв и грунтов солями низкомолекулярных веществ и полимерами. Основными недостатками современных структурообразователей являются высокое содержание солей в поли-комплексной рецептуре, недостаточная водостойкость, низкая агрегирующая способность и неудовлетворительные адгезионные свойства. Частицы почвы, имеющие диаметр 12 мкм, в интервале pH 10,0 – 3,0 суспензия почвы должна быть практически устойчивой, так как величина энергетического барьера значительно превышает энергию теплового движения частиц и составляет 166 kT (pH 10,0), 125 kT (pH 8,0), 63 kT (pH 6,0) и 41kT (pH 5,0). Дальнейшее понижение pH приводит к закономерному уменьшению барьера, который при pH 4,0 составляет 6 kT, а при pH 3,0 полностью исчезает. Таким образом, для фракции суспензии почвы диаметром меньше 12 мкм заметная коагуляция может наблюдаться при pH ≤ 4,0. Легко показать, что более крупные частицы почвы с диаметром 30-33 мкм должны быть практически агрегативно устойчивыми и при pH 3,0, так как энергетический барьер в этом случае составляет 10-15 kT. При взаимодействии самых мелких частиц ($a = 1$ мкм) значения энергетических барьеров составляют: 44 kT (pH 10,0), 28 kT (pH 8,0), 18 kT (pH 6,0), 7 kT (pH 5,0) и 2 kT (pH 4,0) и только для этих частиц при pH $< 4,0$ можно ожидать заметную потерю дисперсией агрегативной устойчивости. Однако, эксперимент показывает, что значительная коагуляция частиц происходит уже при pH 5,0 и непрерывно возрастает по мере понижения pH.

Ключевые слова: структурообразователь; почва; флокуляция; коагуляция; энергия взаимодействия; оптическая плотность; потенциометрическое титрование; электрофоретическая подвижность.