Physicochemical fundamentals of modifying concrete mix and concrete

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Abstract. It is possible to obtain highly efficient construction conglomerates by modifying the structure of cement stone and concrete. Consequently, modification of the concrete structure in this direction is intended to improve the technological properties of the material. By modifying, it is possible to change the kinetics of the growth of physical properties and the final values of the strength of concrete.

Without considering the physical and analytical mechanism for modifying the structure formation of concrete, we note that it is primarily aimed at reducing the amount of water - a mixture in a unit volume of material. However, various studies have proved that in the initial period, the structure formation of the cement stone develops in the optimal direction only at a certain volumetric water content. The limiting reduction in water content makes it difficult to hydrate the binder, limiting the final strength properties of the material. At the same time, the final strength properties of the material are improved with an all-round decrease in the volumetric water content of the mixture. In all cases, the modification of concrete assumes its composition is unchanged from the mixing of the mixture to the final stage of the formation of the concrete structure.

At present, only isolated attempts are known to physically modify concrete, for example, during its evacuation. In this case, the optimal amount of mixing water is introduced into the concrete mixture, which ensures the optimal flow of the binder hydration reaction. The subsequent suction of water, changing the initial composition of concrete, leads to a deep modification of its structure formation. In particular, the density of the material and its strength properties sharply increase. It should be noted that physical modification of concrete leads to a change in the nature of fixation of binder particles.
1. Introduction
As you know, depending on the fixation of particles of interacting substances, two types of bonds can form - weak (secondary) and strong (primary). According to the definition [1], particles form an aggregate if their mutual potential energy \( P = f(r) \) is negative. Therefore, we can conclude that any methods of resource saving and modification of concrete mix and concrete (vacuum, pressing, centrifugation, etc.) will be positive (Fig. 1) if they are aimed at compressing or converging the interacting elements of the material [2].

The main directions of resource conservation in concrete technology

![Image](image_url)

**Figure 1.**
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When two particles of the solid phase approach each other, the surface energy decreases [3] by the value

\[
\Delta \delta = 2 \cdot \delta - \delta_\text{гр} \quad (1)
\]

where \( \delta_\text{гр} \) is the surface energy of the boundary formed between the particles;
\( \delta \) – is the surface energy of the particle,
the force of attraction of these particles to each other will be \( F_k \), and stresses will arise in the contact zone of the particles

\[
P_k \simeq \frac{F_k}{\pi x^2} = \frac{2\Gamma \Delta \delta}{x^2} \quad (2)
\]
where $x$ – is the radius of the contact of the platform, and

$$F_k = \frac{\pi x^2 \Delta \delta}{h} \quad (3)$$

$$h = \frac{x^2}{2\Gamma} \quad (4)$$

Although expressions (3) and (4) were obtained by the author [3] from the assumption of an analogy between the adhesion of solid phase particles and the coalescence of two drops, they express the qualitative direction of structure formation.

The possibility of forming and maintaining stable contacts between particles depends, first of all, on the properties of the surfaces of these particles, the nature of the forces and, especially, their long-range action. With the optimal approach of particles, the cohesion force in the contacts between them will be sufficiently high, which excludes the sedimentation of particles under the action of the force of the gravitational field and the destruction of the structural network as a result of the participation of particles in thermal Brownian motion [4].

In accordance with the theory of condensed phases [5], the formula for determining the adhesion force between spherical particles is:

$$F_C = \frac{2\pi r_1 r_2}{r_1 + r_2} \cdot (2\delta_{13} - \delta_{11}) \quad (5)$$

where $r_1$, and $r_2$ are the radii of the contacting particles;

$\delta_{13}$ – surface tension at the interface solid particle - dispersion medium;

$\delta_{11}$ – Gibbs energy at the boundary of contacting particles (if they are of the same nature).

If $r_1 = r_2$, then

$$F_C = \pi r (2\delta_{13} - \delta_{11}) = \pi r \Delta \delta \quad (6)$$

The resulting expression (6) is similar to (3) obtained by the author [3] by analogy with the behavior of droplets and particles of solid phases. $F_C$ for spherical particles can be calculated knowing the energy of intermolecular interaction of condensed systems [6]:

$$E = \frac{-A}{12H} F_C = \frac{Ar}{12H^2} \quad (7)$$

where $A$ – is the constant of molecular interaction of particles of condensed bodies;

$r$ – is the radius of the particles;

$H$ – is the distance between them.

According to [7], it is proved that at small distances the molecular attraction is $F_C \approx \frac{B}{H^2}$. The last relation is in good agreement with the data [8], which take into account the electromagnetic retardation of dispersion forces for spherical particles:

$$E = \frac{-\pi Br}{12H^2} \quad F_C = \frac{2\pi Br}{H^3} \quad (8)$$

where $B$ – is the constant of molecular interaction taking into account the electromagnetic retardation, if $r_1 \neq r_2$, then

$$r = \frac{r_1 r_2}{r_1 + r_2}$$

According to [9], the electromagnetic retardation of the action of dispersion forces makes a significant contribution to $E$ and $F_C$ at distances $H > 10^{-9}$ m [10], which exceed the absorption wavelengths of atoms and molecules of contacting bodies. Then at atomic (direct) contact of particles $F_C \approx 10^9 \ldots 10^{10}$ H [11], while without taking into account the delay $F_C \approx 10^9 \ldots 10^{12}$ H. According to [12], the value $h_{min} \approx 10^{-9}$m. The probability of particle fixation at a distance of close coagulation increases with an increase in the approach pressure and particle concentration due to a decrease in the potential barrier, while for large particles fixation in a far potential well is more likely [13]. A quantitative measure of this process can be the value of the kinetic energy or compression pressure required to overcome the energy barrier imparted to particles by an external force field. According to [12] for fixation in the position of close coagulation
where $v_0$ is the initial velocity imparted to a particle of radius $r$; 
$m$ is the mass of the particle; 
$\eta$ is the viscosity of the dispersion medium (powder - air).

Since in real systems interactions between particles are of a collective nature, the dispersed systems themselves will obey static laws and a whole spectrum of the above-considered states of particles will be valid for them [14]. The boundary of the transition “coarsely dispersed - microheterogeneous system” is the value of $\delta_c$, determined by the formula:

$$\delta_c \approx \left(\frac{2F_c}{\rho g}\right)^{\frac{1}{3}} \quad (10)$$

formula (10) was obtained by the author of [15] taking into account the fact that one particle has at least two contacts with neighboring particles, which are necessary for the formation of spatial structures with direct atomic contacts.

The interaction energy of the elementary volumes of two spherical particles containing $n$ molecules per unit volume is:

$$E = \int V_1 dV_1 \int V_2 \left(\frac{h^5 \lambda_{ij}}{H^3}\right) \cdot dV_2 \quad (11)$$

where $V_1$ and $V_2$ are the total volumes of spherical particles.

Assuming that the interaction of one molecule will arise with all molecules in the volume of a neighboring particle, after integrating expression (11) we have for spherical particles

$$E = A \frac{r}{H^2} \quad \text{and} \quad F = A \frac{r}{6H^2} \quad (12)$$

where $A$ is the constant of molecular interaction of particles.

Here $n_i$ and $n_j$ are the numbers of molecules of types $i$ and $j$ per unit volume. In the case of a spherical particle and a plane:

$$E = \frac{A \pi r}{6H^2}, \quad F = \frac{4}{3} \pi \left(\frac{Br}{H^2}\right) \quad (14)$$

Since the theory of London does not take into account the electromagnetic delay of interactions, S M Lifshitz [8] developed the theory of molecular interaction of particles on the basis of quantum electrodynamics, taking into account this delay. His conclusions coincide with the experimental data obtained in [9] for spherical particles (8), as well as for a spherical particle and a plane one (15):

$$E = \frac{2}{3} \pi \left(\frac{Br}{H^2}\right), \quad F = \frac{4}{3} \pi \left(\frac{Br}{H^2}\right) \quad (15)$$

Constant $B$ for two molecules is defined as:

$$B = \frac{\pi}{10} n^2 \mu \quad (16)$$

and for particles and condensed bodies as:

$$B = \frac{\pi}{10} \sum_i \sum_j n_i n_j \mu_{ij} \quad (16a)$$

Where $\mu_{ij}$ is a constant characterizing orientational, induction and dispersion interactions, taking into account the delay.

If the contacting surfaces, as in our case, are capable of undergoing deformations, then the energy of their adhesive interaction [16] is calculated as:

$$E = \sum_i F_i = \frac{A}{12} \left(1 + \frac{H+Z}{H} + \left(\frac{H+Z}{H^2}\right)^2\right) - \frac{4\sqrt{2F}}{15K_i} \cdot (H + Z) \quad (17)$$

where $r$ is the particle size; 
$H$ is the distance between the particles; 
$K_i$ characterizes the elastic properties of the particle surface;
\[ Z \cdot \] reflects the deformation of the contacting particles (decrease in the height of the particle, the depth of deepening into the deformable surface), moreover, \( K_i = \frac{1-\varpi_i^2}{E_i^0} \)

where \( \varpi_i \) – Poisson's ratio;
\( E_i^0 \) – Young's modulus.

To study the influence of electric forces on the interaction of particles, let us try to estimate the magnitude of these forces. From the data of the authors of works [10] it follows that with a decrease in the size of particles, their electric charges grow, and, consequently, the electric component of the cohesion force of the particles. According to [17]

\[ F_e = \frac{q^2}{8\pi K^0 X} \left( \frac{\ln\left(\frac{1}{H}\right)}{(0.557+0.5\ln\left(\frac{H}{R}\right))} + (0.557+0.5\ln\left(\frac{H+X}{R}\right)) \right) \]  

(18)

where \( q^2 \) is the effective charge of the particle, which is 20 ... 30% of the total charge;
\( K^0 \) is a constant equal to \( 8.86 \times 10^{-4} \) F / m;
\( X \) is the depth of penetration of the surface charge.

By changing the electrical component, in principle, it is possible to change the adhesion force of particles by decreasing the charge density \( b_s \) :

\[ b_s = e(N_e - Na) \]  

(19)

where \( N_e \) and \( Na \) are the concentrations of donor and acceptor centers on the particle surface, respectively.

Thus, to eliminate the electrical component, it is necessary to bring the interacting surfaces closer to the optimum possible distance.

2. Methods

The main method for modifying the structure of concrete mix and concrete is the use of chemical additives, for example, various surfactants, oligomers and electrolytes. To improve the technological properties of concrete mix and concrete, thermoplastics (latexes, polyvinyl acetate dispersion, etc.), thermosets (phenolic, furyl, epoxy, carbamide resins) and organosilicon compounds are used. SDB, SSB, SNV, SPD, VRP, etc. were used as plasticizing additives. Complex additives with multifunctional action were also widely used. At present, superplasticizers are widely used, the mechanism of action of which is that this plasticizer, interacting with free ions on the surface of cement particles - Ca\(^{2+}\), forms a gel-like compound that eliminates friction between particles and gives a plasticizing effect.

3. Results and discussion

An increase in the adhesion force due to Coulomb forces is observed for particles with an uncompensated charge. The adhesion force in this case is calculated by the method of mirror interaction of the particle charges and the induced charge near the plane [18]:

\[ F_3 = K_3 q^2 \]  

(20)

where \( q \) – is the particle charge;
1 – is the distance between the centers of charges;
\( K \) – is a constant.

The adhesion force component (20) plays an important role precisely for cement, since according to the studies of the authors [19, 20, 21], cement particles have an uncompensated electric charge due to the total negative charge of calcium silicates and positive charges of calcium aluminates, sulfoaluminates and calcium aluminoferrites.

As noted in [22], the presence of a surface charge on cement particles leads to the appearance of a charge opposite in sign and equal in magnitude in the near-surface region. There is a direct relationship between the parameters of the reactivity of cement systems and the electronic characteristics of minerals near the surface zone of cement minerals.
As a component of the adhesion forces, it is also necessary to take into account the capillary, which has the greatest effect on the adhesion of hydrophilic surface particles and the minimum for hydrophobic surface particles. In an air environment, if its humidity exceeds 65%, the capillary component of the adhesion forces of particles prevails over other components; at lower humidity, it will not be so significant.

The force of interaction of two spherical particles of radius r due to capillary forces is:

\[ F_k = 2\pi \delta r \]  
\[ F_k = 4\pi \delta r \]  

(21)

(22)

Taking into account incomplete wetting, introducing the contact angle \( \theta \), equations (21) and (20) transform into (23) and (24):

\[ F_k = 2\pi \delta r \cdot \cos \theta \]  
\[ F_k = 4\pi \delta r \cdot \cos \theta \]  

(23)

(24)

And in the case of direct contact of particles, as well as through the liquid layer, the adhesion forces will decrease with a decrease in the lyophilic properties of the particle surface, that is, with an increase in the angle \( \theta \) [10].

In the general case, the adhesion forces between particles depend on the size of the particles, the properties of their surfaces, external conditions - temperature, impurities and time of contact of particles and, especially, pressure [10].

Determining the contribution of all four components to the magnitude of the adhesion force or autohesion is a task that has not yet been completely solved. Therefore, in each specific case, it is necessary to evaluate the contribution of each component and try to find the resulting force:

\[ \mathbf{F} = \sum \mathbf{F}_i \]  
\[ \sum \mathbf{F}_k = \mathbf{F}_m + \mathbf{F}_e + \mathbf{F}_z + \mathbf{F}_k \]  

(25)

In a liquid (in water), the nature of adhesion forces changes, capillary and electrical forces do not appear, and molecular adhesion forces come first. In general, the force of particle entanglement in a liquid medium decreases, and the intensity of molecular interaction is determined by the thickness of the liquid layer between the particles. Calculations given in [23] show that in aqueous solutions of electrolytes, the molecular interaction between particles is completely screened at \( H = 10^{-6} \) m. At \( H = 10^{-3} \) m

\[ F_m = 0.5F_{\text{max}} \]. For smaller distances, the molecular component of the adhesion force of the particles increases and tends to \( F_{\text{max}} \).

It is necessary to take into account one more important feature of the interaction of particles in liquid media - the presence of a repulsive force associated with the properties of surface layers at the interface between particles and a liquid [24], since with a small thickness of the gap between particles, there is also a wedging pressure in addition to molecular forces attraction. The repulsive forces determine the existence of a potential force barrier (\( F_{\text{max}} \)) that prevents particles from approaching. The force barrier manifests itself at distances comparable to the effective thickness of ionic atmospheres [10]. The forces of interaction between the particles depend not only on the properties of these particles and the interlayer separating them, but also on the external pressing force. If this force is less than or equal to the height of the force barrier (Fig. 2), then the adhesion force is \( F_{\text{min}} \) if the pressing force is greater than \( F_{\text{max}} \), then the adhesion force is \( F_{\text{max}} \).

It is possible to estimate \( F_{\text{min}} \) using [25] for the Brownian motion the expression

\[ \Delta x^2 = \frac{kT}{3\pi \eta r} \cdot t \]  

(26)

where \( \eta \) – is the viscosity of the liquid medium;
\( r \) – is the radius of the particle.
Dependence of the force of interaction between particles on the distance for various external pressing forces.

The interaction energy of particles at a distance $H$ from a flat surface can be represented as [26]

$$E = \frac{\lambda}{\pi kT} \left(0.04083 \left(\frac{10r}{H(H+2r)} + \frac{4H+7r}{(H+2r)^2} - \frac{4H+r}{H^2}\right)\right) \ldots,$$

where $\lambda$ is the wavelength of electrical vibrations of the atom. The value of the interaction energy in the case of the force $F''_{\text{min}}$ should be $\geq kT$, that is, the cohesion force should prevent the free movement of particles.

The splitting pressure $P(H) = -\Delta \mu$ prevents the molecular forces of attraction of the particles; moreover, $\Delta \mu$ is the excess of the chemical potential per unit volume. According to [5], the total wedging effect is equal to:

$$F_{\text{Пакет}} = 2\pi r \int_{H_{\text{min}}}^{H_{\text{max}}} P(H) \cdot dH \quad (28)$$

and according to [8] $P(H) = A H^n$ where $A$ is a constant determined experimentally for $n = 2 \ldots 3$ of each substance.

Then it is possible to calculate the forces acting on the particle due to the wedging pressure [10]:

$$F_{\text{Пакет}} = \frac{2\pi r}{n-1} \left(P_{\text{min}}(H)H_{\text{min}} - P_{\text{max}}(H)H_{\text{max}}\right), \quad (29)$$

where $P(H)_{\text{min}}$ and $P(H)_{\text{max}}$ are the values of the disjoining pressure corresponding to the minimum and maximum thickness of the liquid layer between the spherical particle and the surface.

The wedging pressure is caused by several factors: the molecular (van der Waals) effect of the solid phase on the boundary layer of the liquid and the formation of a double electric layer at the interface (electrical component). This electrical component includes ionic and diffuse:

$$P_{\text{эл}}(H) = P_{\text{n}}(H) + P_{\text{o}}(H) \quad (30)$$

And the total wedging pressure

$$P(H) = P_{\text{эл}}(H) + P_{\text{Пакет}}(H) \quad (31)$$

The electrical component can be represented as a combination of pondermotive forces and osmotic pressure in the layer and volume of the electrolyte [27]:

$$P_{\text{эл}}(H) = P_{\text{n}} + P_{\text{o}} \quad (32)$$
Pondermoto forces are caused by fluctuations of the electric field in the direction perpendicular to the surface of the contacting particles and for a unit surface:

$$F_n = \varepsilon E^2 \frac{8\pi}{\varepsilon^2}$$

where $E$ – is the field strength;
$\varepsilon$ – is the dielectric constant of the medium;
$\varphi$ – is the surface potential;
$H$ – is the distance between them.

Osmotic pressure is associated with the uneven distribution of ions near the boundaries of the particle surfaces:

$$P_o = kT \cdot \sum n_i$$

where $n_i$ – is the number of ions per unit volume of liquid.

Then

$$P(H) = \frac{\varepsilon E^2}{8\pi} + kT \cdot \sum n_i$$

For surfaces with a small amount of charge

$$P_n = \frac{\varepsilon}{8\pi} \frac{\varphi^2}{h_0^2 + C \cdot h^2}$$

and the thickness of the ionic atmosphere

$$h_\alpha = \sqrt{\frac{\varepsilon kT}{4\pi^2Z_i n_i}}$$

where $e$ – is the electron charge;
$Z_I$ – is the electronegativity of the ion;
$C$ – is the concentration of the electrolyte.

At low electrolyte concentrations in the gap and significant thicknesses of the ionic atmosphere, according to the Deryagin-Landau theory [28], the total disjoining pressure is:

$$P(H) = 64n_i k^2 B_0 \exp\left(-\frac{H}{h_\alpha}\right)$$

$$B_0 = \frac{\exp\left(\frac{\varepsilon E^2}{8\pi\varepsilon^2}\varphi_0\right) - 1}{\exp\left(\frac{\varepsilon E^2}{8\pi\varepsilon^2}\varphi_0\right) + 1}$$

where $\varphi_0$ – is the potential of the diffuse electric layer.

If the electrolyte concentration is high enough, then

$$P(H) = \frac{-A}{H^2}$$

At average values of electrolyte concentrations, when the values of $P_o(H)$ and $P_n(H)$ are of the same order, the calculation of the total $P(H)$ is difficult.

By changing the concentration of the electrolyte, it is possible to achieve the disappearance of the force barrier, due to which the adhesion of particles occurs, that is, conditions are created that are favorable for cluster formation.

The electrolyte concentration, above which a force barrier appears (threshold concentration), can be determined according to [9]

$$C_n = C \cdot \left(\frac{\varepsilon^2(kT)^5}{A^2 e^6Z}\right)^f(B)$$

where $C$ – is a constant;
$f(B)$ – a function depending on the asymmetry of the electrolyte, that is, the ratio of the charges of cations and anions;
$A$ – is the constant of intermolecular interaction.

In addition to the above reasons, the wedging pressure can also be caused by the presence of solvation layers, hydration of ions, the effect of an adsorbed liquid monolayer oriented on the surface [29]. The presence of electrolytes in solution, as the authors of [10,29,30] show, affects the cohesion of particles, and, consequently, their clustering, since the cohesion force decreases with decreasing...
electrolyte concentration and depends on the valence of cations. For concentrated solutions, the cohesion of particles decreases with an increase in the valence of cations, and for dilute solutions, it increases.

Sometimes a change in the adhesion force between particles in electrolyte solutions is associated with a change in the electrokinetic potential, which, however, is a function of the thickness of the diffuse layer of adsorbed ions and can only indirectly characterize the change in adhesion forces [31]. Stern proposed a theory according to which the electric double layer consists of adsorption and diffuse, and the nature of electrolyte counterions affects the distribution of ions in it. If counterions have different valences, then the thickness of the diffuse layer and the number of counterions in the adsorption layer depend on the valence of the ions and are determined by electrostatic forces. The higher the valence of counterions, the thinner the diffuse layer and the lower the ε potential. If the ions in the electrolyte have the same valence, then the thickness of the electric double layer and the number of counterions in the diffuse layer depend on the polarizability and hydration capacity of the ions: the greater the polarizability and the less hydration, the smaller this thickness [32, 33]. The charge of the adsorption layer according to Stern [31]:

$$q_{ads} = F_z (\Gamma^+(\theta^+)+\Gamma^-(\theta^-)) + \frac{1}{1+\frac{C^\infty}{\exp\left(F_z \varphi \delta / RT\right)}} - \frac{1}{1+\frac{C^\infty}{\exp\left(\theta^+(\theta^-)+F_z \varphi \delta / RT\right)}}$$

(42)

where \(\Gamma\) – is the number of ions of different signs adsorbed by a unit of surface; 
\(\theta\) – specific adsorption potential for (+) and (-) ions; 
\(F\) – is the Faraday number; 
\(Z\) – valence; 
\(C^\infty\) – is the concentration of the electrolyte at an infinite distance from the solid phase; 
\(F_z \varphi \delta\) – electrical adsorption potential.

Diffuse layer charge:

$$Q_{diff} = \frac{S\pi T}{2\pi} C^\infty \left(\exp\left(F_z \varphi \delta / 2RT\right) - \exp\left(-\frac{F_z \varphi \delta}{2RT}\right)\right)$$

(43)

The charge on a solid surface: 
$$q = q_{ads} + q_{diff}.$$

The deformation of the electric double layer can occur not only under the action of an external electric field, but also under the action of convective flows of electrolyte, gravitational field, field of various mechanical influences. Upon deformation of the electric double layer, polarization of the ionic layers occurs, leading to the appearance of long-range attractive forces between the induced dipoles, which plays an essential role in structure formation.

4. Conclusions
Based on the above, it is necessary to take into account the following aspects of the influence on the change in the energy of cement systems:
- interaction of particles at the molecular level with the resultant force GM;
- electrical interaction of particles, including due to the mirror effect with the forces Fe and Fa;
- interaction of particles taking into account the capillary forces Pcap;
- the number of particles per unit volume, that is, their concentration;
- the nature of the dispersion medium;
- the presence of an external force field.

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