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The Combined Action of Acoustic and Electric Fields to Nanofluids for Concrete

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ABSTRACT:A technology has been developed for the integrated activation of natural water, modified with SiO_2 nanoparticles 40 nm in size for the purpose of using it for concrete-cement mixtures and for the production of earthquake-resistant high-strength concrete. These physical conditions allow achieving an increase in the degree of compression by 20% for the tested samples in comparison with the comparison sample. It is shown that the fundamental role in the activation mechanism is that, as a result of the reaction of hydrogen to hydrogen compounds, hydrogen bonds between oxygen and oxygen are formed, and activation occurs during the intensification of these processes. In addition to standard components, silicon dioxide was used as modifying additives, which is a product of gas treatment plants in the technical silica production. In this case, a decrease in filler is crucial. Electrochemical (voltage 15 V, current 4.5 mA/cm^2, steel electrodes) and synchronous acoustic field (22 kHz; 4,0 Wt/sm^2) was combined. A comparison sample was carried out similarly, but without modifications and activation of mixing water. The tests were carried out on samples made of concrete mix with a size of 40x40x160 mm^3 for 28 days after they were poured according to standard methods.

I. INTRODUCTION

The bottom-up growth of networks is a fascinating approach for the development of new functional nanomaterials with tunable mechanical and thermophysical properties. Hereby, water molecules are the building blocks, which can be activated by external mechanic and electromagnetic field conditions and by add to solution with nanoparticles. By the proper design of the precursor molecules and by controlling the parameters such as concentrations, and substrate temperature or irradiation, chemical reactions can be induced.

It is most effective to increase the concrete performance characteristics associated with strength and seismic resistance, activation of mixing water with the help of acoustic, electrical or electromagnetic fields that contribute to the stabilization of the structural and technological characteristics of the product. Ultrasonic and electromagnetic waves used for activation significantly increase the uniformity of the resulting product, contributing to its seismic, environmental and climatic sustainability, which are relevant in the construction of supports for solar cell mounts. Such methods of physical processing of mixing water are being intensively developed along with the chemical modification of Portland cement systems by introducing organic and mineral additives into them, taking into account the fact that the water present in the concrete mixture is its active component. In the process of the formation of chemical bonds and adsorption boundaries, water determines the hydration and its parameters during the formation and maturation of

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concrete stone, including the rate of hardening and final strength, which ultimately sets the compressive strength and seismic resistance of building structures. Since the activity of water depends on its energy state, various physical effects on its structure intensify the processes occurring with its participation. In particular, the activation of water by acoustic or electric fields causes not only a change in its ionic composition, but also leads to an increase in the activity of hydrogen ions and the structuring of hydrogen bonds. The indicated physical effects impart high chemical and hydration activity to water. At the same time, concrete systems obtained using activated mixing water acquire predetermined technological properties and physicochemical features. At the present work, we have studied the effect of acoustic and electric fields to nanofluids in order to develop new approaches based on the use of water dispersible compositions.

II. RELATED WORK

In our previous research [1-5] we shown complex chemical relaxation spectra in different solutions in utrasonic field. Therefore, there are still considerable scientific challenges at play, making it a current hot topic of soft condensed matter. Cluster and clathrate models imply the additional formation of pulsating cavities of various sizes, which may contain in their internal volume both individual wandering monomolecules of the water itself and guest molecules that increase the stability of the structure [6-8]. Hydration processes can be enhanced by the addition of structuring agents, for example, such as SiO_2 nanoparticles, the sizes and concentration of which are interdependent.

Experimental and theoretical studies of the structure and properties of water under the influence of external influences, providing the possibility of its targeted regulation, are of great practical importance when used in production and technological processes associated with its hydrating abilities. Despite the existence of various water models, the main distinguishing feature of each of them is the experimentally established fact of the presence of a complex branched network of hydrogen bonds in which short-range order is present. These processes are especially relevant when using activated water modified by silicon dioxide nanoparticles to prepare concrete mix when pouring the foundations of constructed industrial and civil construction objects, as well as numerous products from concrete, foam concrete, reinforced concrete. These structures must correspond, first of all, to compressive strength, which provides resistance to all types of load and take into account soil features [9]. To achieve this goal in the manufacture of concrete mixtures, it is envisaged to use both various composite additives to modify the most important operational properties [10], and the use of complex methods for activating mixing water to ensure hardening and seismic resistance of the final product. To improve the operational parameters of concrete, various technological methods are now widely used: ultrasonic, electrochemical, electromagnetic activation of mixing water, the use of effective modifying chemical additives, as well as the selection of the dispersed range of the aggregate mixture.

III. PHYSICAL METHODS OF ACTIVATING MIXING WATER AND ITS STRUCTURING EFFECT

The effect on the ionic composition of the water used for mixing can control the operational properties of the resulting concrete stone. To mix concrete mortar used in construction, natural water with different ionic composition is usually used, which determines the behaviour of its structure as a result of the influence of the acoustic field, electrochemical exposure or modification with various additives [8,12-16]. In the case of electrochemical activation of mixing water using steel electrodes, there is a high probability of the formation of crystals of iron oxide-hydroxide compounds having sizes from 1 to 100 nm, which are unstable, and are stabilized through the formation and coagulation of the formed micellar structures of variable composition. When interacting with electric and acoustic fields, these dispersed structures can be active crystallization centres during hydration, further affecting the structure and mechanical properties of cement stone. When using an aluminium anode in the volume of electrochemical activation, the thermodynamic properties of the aluminium-water system, hydrogen is reduced at the cathode and the near-cathode solution is alkalized. In an alkaline medium, aluminium hydroxide dissolves to form AlO_2^- aluminate. The accumulation of dispersed particles of hydrargillite at the time of aluminium oxidation provides the formation of micellar structures that stabilize these particles, which provides the possibility of electrochemical activation of water. The flow of electric current through water is accompanied by processes of displacement of the cathode potential into a more negative range of values. In this case, the restoration of formations with a positive charge is realized, the features of which are determined by the charge, concentration and structure of the arising associates (ions, micelles, clusters, etc.). Certain features of the physical properties of water, caused by a network of hydrogen bonds between adjacent hydrogen and oxygen atoms, create the prerequisites for the functioning of clusters in an aqueous solution, the

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intensification of which is initiated by the parameters of its activation procedure [17,18]. The development of molecular and structural-chemical concepts made it possible to explain the ability of water molecules to form hydrogen bonds and substantiate the role of bound water in the processes of hydration of substances and their physicochemical properties and final parameters.

In terms of energy, the hydrogen bond occupies an intermediate position between chemical covalent or ionic bonds and intermolecular Van der Waals interactions in the liquid state. The energy of the hydrogen bond is 5–10 kcal/mol, while the energy of the covalent O–H bond in water is 109 kcal/mol [19,20]. Hydrogen bonds in the liquid state constantly arise and disappear as a result of thermal fluctuations. Another important property of the hydrogen bond is the cooperativity of the interaction. The formation of one hydrogen bond contributes to the formation of the next hydrogen bond, which, in turn, promotes the formation of the next hydrogen bond, etc. Molecules of water in a liquid state under normal conditions (1 atm, 22 °C) are mobile and capable of oscillating movements, rotations around its axis, as well as chaotic and directional movements, due to which individual molecules can "jump" from one place to another in volume water through cooperative interactions. As a result, autoprotolysis is possible in aqueous solutions, i.e., detachment of the H^+ proton from one water molecule with subsequent movement and addition of H^+ to the neighbouringH_2O molecule with the formation of hydroxonium ions. This leads to the fact that water should be considered as an associated liquid from a set of H_2O molecules connected by hydrogen bonds and weak intermolecular van der Waals forces. An example of such an associate is a water dimer: $H_2O = H_2O + HOH$. Hydrogen bonds are easily destroyed and quickly re-formed, which makes the water structure unstable and variable over time [20]. This process leads to heterogeneities in the structure of water, which characterizes water as an associated heterogeneous two-phase liquid with a short-range order, i.e., ordering in the mutual arrangement of atoms and molecules, which repeats in the first coordination sphere - at distances comparable with the distances between atoms. The random movement of liquid molecules leads to a continuous change in the distances between them. The statistical nature of the ordered arrangement of liquid molecules leads to fluctuations - continuously occurring deviations not only from the average density, but also from the average orientation, since liquid molecules are able to form groups in which a certain orientation prevails. Moreover, the smaller the magnitude of these deviations, the more often they occur. Hydrogen bonds are spatially oriented. Since each H_2O molecule has four centers of hydrogen bonding (two non-separated electron pairs at the oxygen atom and two uncompensated positive charges at the hydrogen atoms), one H_2O molecule in the condensed state is capable of forming hydrogen bonds with four H_2O molecules, which leads to the formation of a tetrahedral crystalline structure organized in clusters. Clusters are fairly stable and can be isolated for a short time. It has been established that charged ions stabilize clusters. Therefore, it is possible to subdivide clusters into charged cluster ions and neutral clusters. According to water studies by IR and femtosecond laser spectroscopy, as well as mass spectrometry, clusters containing 20 H_2O molecules and a proton in the composition of the hydroxonium ion H_3O^+ form cluster ions of the composition $(H_2O)_{20}H_3O^+$ or $(H_2O)_{21}H^+$, which turned out to be the most stable. It is assumed that the stability of such cluster ions is explained by a special clathrate ion, i.e. having a cavity with a structure in which 20 water molecules form a 12-sided polygon - a dodecahedron, in the cavities of which there is a proton in the form of a hydroxonium ion H_3O^+. Around the modifying nanoparticles present in the water: ions, molecules, various types of aggregates, hydration shells are formed. According to the model of hydrophobic hydration, water molecules form a clathrate structure around dissolved molecules. The stability of clathrates depends on the nature of the guest molecule, concentration, temperature, pressure. The chemical potential of a modifying nanoparticle, which can be considered within the framework of nanothermodynamics [21] as a supramolecule consisting of a set of {n} structural units (n_1 structural units of the first grade, n_2 structural units of the second grade, etc.), using the result of statistical mechanics for ordinary molecules (as given in [21]) has the form:

$$\mu_{\{n\}} = G^0_{\{n\}} + kT \ln(c_{\{n\}}\Lambda^3_{\{n\}} f_{\{n\}})$$

where $\mu_{\{n\}}$ is the chemical potential of the supramolecule, $G^0_{\{n\}}$ is the Gibbs energy of the supramolecule {n} in this medium, provided that the center of mass is at rest and there are no other supramolecules; k is the Boltzmann constant, T is the temperature; $c_{\{n\}}$ is the concentration of supramolecules; $\Lambda_{\{n\}}$ is the average de Broglie wavelength and $f_{\{n\}}$ is the activity coefficient of supramolecules, reflecting their interaction. The average de Broglie wavelength of the supramolecule is determined by:

$$\Lambda_{\{n\}} \equiv h(2\pi m_{\{n\}}kT)^{-1/2}$$

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where h is Planck's constant; $m_{\{n\}}$ is the mass of the supramolecule, consisting of the masses of the structural units m_i

$$m_{\{n\}} = \Sigma m_i n_i$$

The quantity $G^0_{\{n\}}$contains accounting for the interaction of the supramolecule with the medium; if we include in $G^0_{\{n\}}$the activity coefficient $f_{\{n\}}$ in the composition of the quantity $kT \ln f_{\{n\}}$, the expression for the chemical potential takes the form:

$$\mu_{\{n\}} = G^0_{\{n\}} + kT \ln(c_{\{n\}}\Lambda^3_{\{n\}})$$

where $G^0_{\{n\}}$ already takes into account the interaction of the supramolecule not only with the medium, but also with other supramolecules. The properties of self-organized systems in a liquid nanomatrix can be controlled by modifying it with various types of nanoparticles. These can be fullerenes, carbon nanotubes, SiO_2 nanoparticles, etc., which are involved in the self-organization process as hydrophobic objects of Van der Waals interactions and provide the whole variety of ongoing relaxation processes [22] and physicochemical processes, such as hydration during the concrete curing process.

IV. RESULTS AND DISCUSSION

For the preparation of the concrete mixture used Portland cement grade M500 D0 according to State standard GOST 31108-2003. The figure 500 means that a cement stone made from such a powder is able to withstand a maximum brand load of up to 500 kg per 1 cm^3. Another part of the cipher is the letter "D" with figures from 0 to 20. They reflect the number of additives introduced into the composition of the material in percent. In addition to the standard constituents, nanosized (40 nm) silicon dioxide was used as a modifying additive. In this case, the dispersion of the filler plays a decisive role, the increase of which allows to reduce the optimal consumption of the additive. As mixing water, ordinary tap water was used, subjected to complex activation by an acoustic field (frequency 22 kHz, power 4.0 W/cm^2) and electrochemical treatment (voltage 15 V, current density 4.5 mA / cm^2, steel electrodes). The comparison sample was made similarly, but without modification and activation of mixing water. The tests were carried out on samples made of concrete mix measuring 40x40x160 mm^3 on day 28 after they were poured using standard methods: “Concretes. Methods for determining the strength of the control samples (State standard GOST 10180-90)"; “Concrete. Methods for determining the density (State standard GOST 12730.1-78.)"; "Transient Hot Bridge method for determining thermal conductivity, thermal diffusivity and heat capacity."

The results are shown in Table 1.

Table 1
Physical parameters of the developed concrete composition

№	Operational parameters	Designed composition	Comparison sample
1	Compressive strength, MPa (kgf / cm^2)	60,9 (621)	50,6 (516)
2	Density, kg/m^3	1572	1391
3	Thermal conductivity, W/(m · K)	0,630	0,788
4	Specific heat (at 25 °C), kJ/(kg · K)	1,321	1,174

The results show that the developed concrete composition in terms of parameters 2-4 is close to the class of high-strength lightweight concrete [23] while the compressive strength is more than 20% higher than the strength of the comparison sample, but 40% less than the strength of the sample, considered in [24] - especially heavy radiation-resistant high-strength concrete, which is explained by the absence in the developed sample of a moisture-retaining heavy additive in the form of waste products of ferrous metallurgy. Moreover, samples from the developed composition are characterized by values of compressive strength of 621 kgf/cm^2, which, according to the requirements of State standard GOST 10178-85, classifies them as guaranteed grade 600, while the reference sample has grade 500, which indicates the possibility of increasing the grade of concrete using activated by a developed method of mixing water.

V. CONCLUSIONS

We have developed new technological approaches based on the use of water dispersible compositions [25], which currently successfully displace products based on organic solvents, being environmentally safer both in production and in operation. The use of modifications by various nanoparticles to control the behavior of the nanofluid [26-29], what the mixing water becomes in this process, is most effective while using the electrochemical activation method and the influence of acoustic fields on its structure. In the process of further technological modification using silicon dioxide, the material acquires predetermined properties necessary in practical application. In this direction, the beneficial properties of mineral and organic components that function as blowing agents, dispersants, stabilizers, binders are optimally combined. In the process of testing concrete mixtures, the combined method of activating mixing water, combining ultrasonic and electrochemical methods of exposure, proved to be most effective. Due to the synergistic processes in the solvent, the efficiency of the operational characteristics, expressed in the ability to stabilize, hydrate, strengthen properties, is significantly increased. The combination of these parameters makes it possible to create modified cement-concrete solutions of increased strength and seismic resistance. This indicates the prospects of the developed technology for use in the manufacture of concrete structures with a wide profile including the special physical properties.

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