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PHYSICAL RELAXATION AND STRUCTURE FORMATION OF CONCRETE

Problem statement. Key problems of rheology at the physical stage of concrete structure formation are considered.

Results and conclusions. Connection between behavior of dry and wet granular systems and their fractal structure is discussed. Fundamental features of density relaxation of dry granular systems as well as the loss of their stability under internal surface wetting are examined. The main factors influencing the rheological properties of concrete mixes are determined. The methods of management of kinetics and concrete structure formation directions are suggested.

Keywords: rheology, concrete, structure, granular systems, stability, relaxation, kinetics.

Introduction

The main challenge of materials technology is to mould materials with prescribed properties. For this purpose, first the desired structure is to be determined and obtained with the use of one of other technological techniques. We restrict the discussion to the problem of obtaining the wanted structures at the stage when physical process predominate over the chemical ones (i.e. early structure formation). Although our study has been to a certain extent restricted to that, there are lots of crucial issues to examine.

First and foremost, it should be stressed that structure formation crucially depends on the state of aggregate of the basic components. The gaseous state is the most suitable favourable for a material synthesis. In the above state, molecules of an initial material are able to move freely and form various chemical bonds which are essential to the ultimate structure of a material. Among other kinds of raw materials, natural gas is of particular interest for the development

of similar techniques. Incineration of natural gas as fuel undermines large-tonnage production of unique materials in the future. Nonetheless, gas is extensively used for polymers synthesis nowadays.

Polymer materials along with timber are a promising source for Russia possessing unique gas deposits. However, polymer synthesis is a separate complex issue, and thus will not be dealt with.

Another state of aggregation, which is also very convenient for materials production due to the molecular mobility, is a liquid state. Here, metals and glasses are getting a leading position and they are making up the growing proportion in the overall building material structure. Besides, there are a lot of interesting and practically vital issues of polycrystalline solids and various amorphous structure formation calling for attention.

Last but not the least, there is a broad class of concretes formed on the basis of various types of fillers and binding materials that make up the ultimate material matrix. The merits of concrete as a building material are versatile. Among them, we should emphasize high durability, capacity to make up large-scale constructions of various forms and reclamation of natural and anthropogenic solid materials. Some features of concrete structure formation hamper its full application. Concrete, in particular, is largely made up of rather inert granular materials such as sand and crushed rock. In the Earth's field of gravity without any exposure they form masses where there is practically neither movement nor mixing of the components.

1. The granular systems general characteristics

Granular materials are structurally simple. They consist of a large number of macroscopic particles. If these particles are not binding, there are mainly act-on-contact pushing forces between them. If particles are dry, any internal fields different from contact forces may be considered small. Despite the structural simplicity, the behavior of granular systems is quite different from the other matter forms such as solid bodies, liquids and gases which may be considered as additional states of matter. One of the distinctive features of the granular systems is that interaction between granules is dissipative due to the static friction and non-elastic collision.

The flow of friable granular medium can to a certain extent be likened to a liquid. Nonetheless, here, unlike liquid or gas, ordinary thermodynamic temperature T is of no importance, since the energy of granules is $E >> kT$. Hence, it is impossible to develop the

granular medium thermodynamics using the analogy with liquid or gas. The granular materials show unusual behavior even at rest. Thus, sand in a container presses the bottom with the force $F < \rho Shg$ due rubbing particles transferring some of the force onto the container walls. The tension inside the medium is extremely heterogeneous; the tension along the chains formed by it is the largest. These chains play a large role in many granular medium properties, in sound distribution, for example. Force distribution in the particles is $P(f) = C\exp(-f / f_0)$. Such a behavior may be explained within a simple model where the masses located in a random grid distribute their weights randomly and on average evenly onto all the particles of the lower layer. This model in some ordinary cases is really in good agreement with the experimental data and numerical simulation results.

2. Relaxation of friable systems density

One of the most important phenomena for the granular mediums physics is their ordering. For a random ensemble of mediums, the degree of packing ranges from $\eta = 0.55$ to $\eta = 0.64$. Due to the static friction, metastable states between these two limits can be present for a considerable amount of time, since the thermal energy *kT* can be neglected. Density may thus change only as a result of external disturbances such as vibration or wetting. The investigations show that granular systems relaxation occurs logarithmically slowly. The most suitable explanation for this phenomena is presumably based on the notion of fractal structural features and of the system energy pattern. In order to determine whether an object is fractal, it is necessary to find its fractal dimensionality *D*.

The easiest way to do it is a cell method where a structure under study is thoroughly covered with disjoint cubes of Euclidean space of the size ΔR. A number of such cubes is then calculated and the power law between their number and scale is determined:

$$N\ \Delta R\ \sim \Delta R^{-D}, \qquad (1)$$

where *D* is a fractal dimensionality. The relation (1) can be well applied to real objects only in a certain range of scales. Fig. 1 presents how a typical fractal line looks like.

A lot of porous structures have multifractal properties. The geometric meaning of that is that a multifractal object is a system divided into many parts, and yet it also has a fractal structure but with a different dimensionality. It was shown that in case of adsorption in porous

mediums when a monocular covering of a surface is performed, the concentration in a pattern volume is

$$n \sim \Delta R^{-D}, \tag{2}$$

where $D < 3$. In experiments with a porous carbon adsorption $2.16 < D < 2.97$ for different gangues. The results for the fractal dimensionality obtained by the adsorption method are in good agreement with the experimental data on the liquid flow. The fractal approach is applied for various objects with a strongly developed surface.

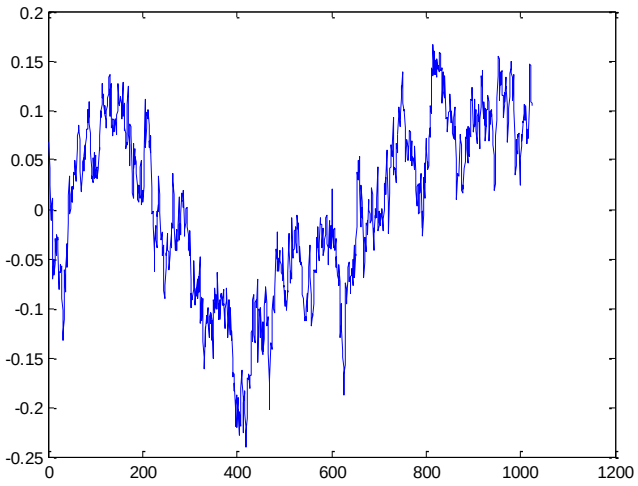

Fig. 1. Fractal line

For some fibre-granular materials such as sand, cement, and PVC granules the processes of density relaxation were studied under the influence of vibration. A random structure defined by the presence of particle arches. Force chains, which in their static state perceive a loading from upper layers, form around these arches. The particles outside the arches do not transform the common tension in the system. The newly formed porous space is of fractal character [1]. The whole system is a group of randomly formed void clusters. The effect of vibration destroys the balance of the system, thus changing its structure. A theoretical model for relaxation under vibration may be built on the basis of fractal kinetics of hierarchically subordinated systems which is illustrated in [2] assuming that a variety of void clusters in the system have a power-series (fractal) distribution in the sizes R:

$$\omega(R) \sim R^{-D}. \tag{3}$$

Let us assume that relaxation at each scale has an exponential form

$$S(R,t) = \exp(-t/\tau), \tag{4}$$

and the relation between the time of relaxation and disturbance energy is determined by the Boltzmann law

$$\tau = \tau_0 \exp(E/T), \tag{5}$$

where τ_0 is a typical time; E is a disturbance energy of a pore rearrangement; T is an average chaotic motion energy which for vibratory disturbance is equal to a granule average vibrational energy.

If we consider the dependence relation between disturbance energy on the value R to be a power-series one as well, we get the following:

$$E(R) = Q_a R^a, \tag{6}$$

a decrease of a void cluster average size with the lapse of time is described by the ensemble average:

$$\frac{\bar{R}(t)}{R_0} = \sum_R \omega(R) S(R,t) = \sum_R R^{-D} \exp\ -t/\tau_0 \exp(Q_a R^a / T)\ . \tag{7}$$

The asymptotic evaluation of the expression (7) offers the following:

$$\rho\ t\ = \rho_f - \frac{\Delta\rho_\infty}{1 + B\ln\left[1 + \frac{t}{\tau}\right]}, \tag{8}$$

where $\rho(t)$ is a current density value; ρ_f is the ultimate density; $\Delta\rho_\infty$ is a total increase in density caused by relaxation; τ is a typical relaxation time constant. The function (8) is shown in Fig. 2.

The results of the quantitative analysis [3] show that the dependence presented is in the best agreement with the experimental data obtained. The system relaxation at moderate vibration accelerations $a \sim g$ is thus described by the reciprocal logarithmic law according with the fractal notions of granular materials. Laws for coarsely and finely dispersed materials

relaxation are identical, which allows to assume that they are independent from force correlations between the particles in a wide range of the system parameters.

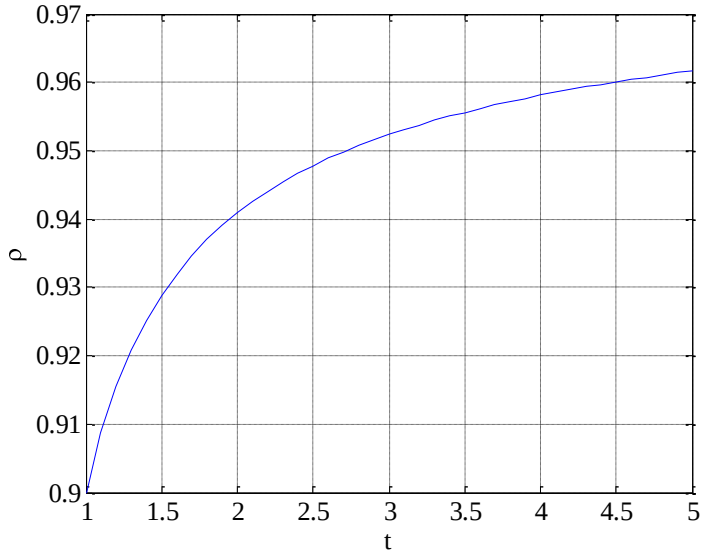

Fig. 2. The dependence of granular system density on time during vibration

3. Topological properties of the porous medium and its durability

Porous bodies are quite complex in terms of their topological properties. The Betti numbers are used for characterizing porous space cohesion. Topology fundamental law states that two structures are topologically identical if and only if all the Betti numbers are equal. The Betti numbers β_n can be found for a given structure but their precise estimation requires a profound knowledge of topology. With a practical aim we will restrict ourselves to the first three Betti numbers.

The zero Betti number β_0 points to the number of isolated clusters in a structure. To put it this another way, β_0 is a number of separate components making up a structure. For instance, the granular space of a sandstone bar has $\beta_0 = 1$. Thus, the assessment of β_0 may testify that a structure includes some isolated porosities. The first Betti number β_1 is a number of holes crossing the structure: $\beta = E - N_v + 1$, where E is a number of faces, N_v is a number of vertexes of the system equal to a number of pores. The number β_1 coincides with the maximum number of non-intersecting closed curves which may be drawn on a surface without dividing it. If torus, for example, is cut along a closed curve, the resulted figure

deforms into a cylinder (connected set). Besides, if we cut a cylinder, it will split into two isolated clusters, hence the Betti number for a torus is equal to one. The characteristics of a surface type G normalized for a unit volume is frequently used. $G_V = z/2$ is a half the coordination number, i.e. the average number of the surrounding pores with which a given pore has an immediate connection with. The first Betti number is also a measure of a set of number of independent ways within the structure.

Finally, the second Betti number β_2 is a measure of a number of the sides in a system, i.e. sides of the inner surface. For instance, the Moebius band is a one-side surface with $\beta_2 = 1$. After being moved along the closed contour, the normal vector to such a surface hits the same point and turns out to be directly opposed to the initial position.

The topological and geometrical forms are connected by the Gauss-Bonnet theorem. The Gaussian surface curvature is

$$\langle k_G \rangle = \int_Q k_G ds, \tag{9}$$

where the Gaussian curvature $k_G = k_1 k_2$ and k_1 and k_2 are maximum and minimum curvature at a given point of a surface. According to the Gauss-Bonnet theorem [4], $\langle k_G \rangle = 4\pi(1 - G_V)$. For a porous surface we get $G_V >> 1$ and $\langle k_G \rangle < 1$. This may be fulfilled only for porous systems with predominately saddle-like structure of inner surfaces as shown in Fig. 3.

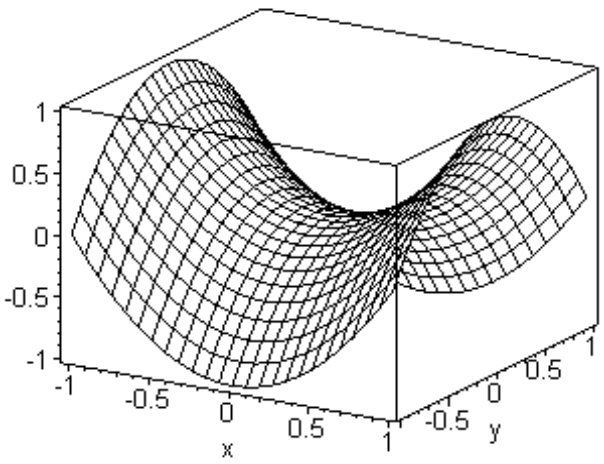

Fig. 3. The characteristic saddle-like surface

Such a structure of a granular system inner surface results in the fact that as thin continuous water film wetting its inner surface increases, unbalanced binding effect undermining the inner mechanical durability. As a consequence, the system thickens [3]. This phenomena is of significant meaning to the building material science.

4. The cluster structure of aqueous suspension hydrodynamic flows

In order to account for granular mediums flows, a granular hydrodynamics is developed (despite the fact that there is no water here). The most prominent feature of processes in both dry and damp granular mediums is particles clusterization as a result of their non-elastic collisions. Actual phenomena in granular systems can be rather complex. They can include heat and mass transfer, thermodynamic phase change and may proceed with the participation of different forces such as viscosity and capillarity.

Granular materials show various behavior patterns, which may be typical both of solid bodies and of liquids and gases. Wetted systems rheology consisting of solid, liquid and gas phases is determined by interfacial and interparticle interaction. In the classical rheology the relation between the stress and deformation is are found experimentally. This relation in disperse systems, unlike Newtonian-like ones, is of nonlinear character.

The properties of these structures to a great extent depend on a form, size, the particles' mutual arrangement, their concentration and thickness of liquid interlayers or interparticle distance separating them [6, 7].

Thus, for example, the linear relation between pressure differences and deformation or a velocity gradient is affected. The classical rheology states but does not reveal the fact, thereby microrheological approach to disperse systems description is of particular interest.

The microscopic rheology is a result of a dynamics manifestation in small scopes. Atomic and molecular properties of granular systems cannot directly impact the rheological properties. In actual viscous systems used, for one, in engineering, they show themselves as fairly large adjustments forming disperse and large particles of fillers. Interaction forces in such heterogeneous systems are thus realized at the mesolevel. A detailed physical mechanism of rheological processes in watered granular systems and emulsions is now blurred. Thus simplified models providing explanation for the nature of viscosity of such systems are of interest to us.

If fairly large particles of a more than a micron are examined, the Brownian motion can be neglected. Flows for the most important applications are low-speed ones, therefore turbulent effects are not present, while aggregation processes are reversible. Aggregation brings about fractal clusters formation [8]. Making an account of suspensions fluidity, we assume that suspension particles produce aggregates in the form of fractal clusters mainly at the expense of capillary-film interaction. Fractal clusters behave similarly to compact hydrodynamic spheres with a hydrodynamic radius equal to the radius of inertia. As this takes place, the liquid phase is partly pulled into a cluster facilitating its compaction. The fractal dimensionality of the clusters of $2 < D < 2.3$ is less than the corresponding Euclidean space dimensionality, i.e. the clusters are rather friable.

In order to clarify the models, the clusters fractal peculiarities and also the possibilities for the destruction of the clusters in a shearing flow should be taken into consideration. While obtaining a cluster equilibrium radius from the medium field approximation, viscosity of the effective environment surrounding a cluster is presumed to coincide with that of a suspension. We may resort to the law for suspensions viscosity of randomly distributed rigid spheres and then substitute the volume of the spheres by the volume of the aggregates thus obtaining an analytical viscosity-shift stress ratio:

$$\frac{\mu_A(\tau)}{\mu_0} = \frac{(1-\varphi_A)}{(1-\varphi_A/\varphi^*)^2}, \tag{10}$$

where

$$\varphi_A \approx \varphi^* \left[1 + \left(\frac{\tau^*}{\tau} \right)^m \right]^{3-D}, \tag{11}$$

$\mu_A(\tau)$ is the effective cluster suspension viscosity; φ_A is the effective clusters concentration; φ^* is the critical clusters concentration; τ^* is the limit stress of a shift when the clusters destroy; D is the clusters fractal dimensionality.

Shift flows, which bring about moments of forces partially destroying the clusters, have the most drastic influence on their size. The effective radius of the aggregates and also a suspension viscosity thereby become the shift stress and flow velocity function. The liquid subsequently ceases to be a newtonian one. The aggregates selective destruction and resetting in a flow may cause the generation of structures with a lower porosity at subsequent growth

stages and also bring about the formation of more dense structures at higher shear rates. The correlation between a hydrodynamic and effective cluster radiuses is essential to the examination of suspension rheology. Fractal clusters with a strong interaction inside the aggregates and having a small viscous dissipation in a liquid phase behave similarly to particles of the solid phase in a viscous liquid.

Theoretical and experimental investigations of relaxation and shift dynamics in granular systems exhibit a definite versatility of their behavior at a mesolevel. A fractal cluster structure plays the crucial role in the properties of both dry and watered systems. The properties of clusters, their destruction and resetting account for the peculiarities of dense granular systems behavior.

As far as the practical applications are concerned, the description of viscous suspensions flow is of fundamental importance. Here, it is easy to obtain the results if the suspension viscosity is determined by a power law from the shear rate. Hence we can write the following for the pressure differences $p(x)$ during a liquid flow in a tube at an area with an infinitely small length dx:

$$[p(x+dx)-p(x)]\pi r^2 = \mu_o 2\pi r dx\left(\frac{dv}{dr}\right)^n. \qquad (12)$$

Rearrangement of Eq. (12) gives

$$\left(\frac{\partial p}{\partial x}\frac{r}{2\mu_0}\right)^{1/n} = \frac{dv}{dr}, \qquad (13)$$

where pressure gradient is considered to be a constant one along the tube. Eq. (13) is at once integrated. As a result, we get the generalized Poiseuille formula for speed distribution along the tube radius:

$$v(r) = \left(\frac{\partial p}{\partial x}\frac{1}{2\mu_0}\right)^{1/n}\left(\frac{r^{1/n+1}}{1/n+1} - \frac{R^{1/n+1}}{1/n+1}\right). \qquad (14)$$

A change in the nature of the relation between a flow and a pressure difference in concretes as compared to ordinary liquids have a strong effect on the possibilities for their long-distance transportation along tubes.

5. Structure formation control

As various as structure formation mechanisms are, they are all of statistical nature. In view of this, the natural way of describing the structure evolution is a master equation for f_i distribution function by locally stable states:

$$\frac{\partial f_i}{\partial t} = \sum_j (w_{ij} f_j - w_{ji} f) . \quad (15)$$

Here, $w_{ij} = \exp(-E_{ij} / T)$ are kinetic coefficients having the value of shear rates from the state with the index *j* into the state with the index *i*, E_{ij} is the energy of a separating barrier in the shift direction.

In cases of kinetic coefficients symmetry in the indexes *i*, *j* relaxation and a transition into a more stable structure respectively do not take place. Such a form of kinetic coefficients is typical of systems with a random energy pattern with approximately similar potential barriers for the transfer of the states in the forward and backward direction, as shown in Fig. 4.

If an energy pattern tends to decrease as shown in Fig. 5, relaxation will occur in a system, i.e. a gradual stochastic transition from states with a higher energy into states with a lower energy.

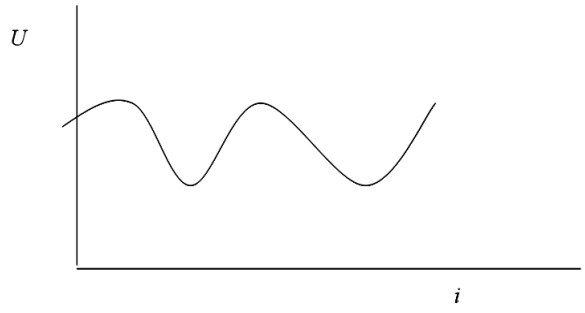

Fig 4. Potential pattern without a gradient

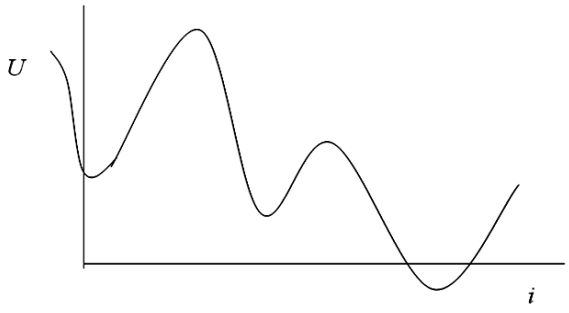

Fig 5. Potential pattern with a gradient

Since spontaneous relaxation in concretes is almost impossible, in various technological processes it is often stimulated by vibration or a forced mixing that brings about chaotic changes in a concrete structure. The highlighted general properties of the relation between the relaxation rate and an energy surface shape show that artificial changing of kinetic coefficients is needed for the control of the direction and the rate of structure formation. The overall speed of the processes may be increased at the expense of increasing all of the kinetic

coefficients. This can be achieved, for example, by increasing the vibration level. However, such a procedure results in a more steady system mixing in all the states possible, which is not always desired if we are seeking to create specified mesostructures inside the material.

Grouping energy structures into certain energy zones provides another way of controlling the process of structure formation. Then, in case of the conservation of the general potential difference of an energy decrease we get an increase in the height of potential barriers for backward transitions with an energy increase. In case of the conservation of the potential barriers height we get a rise for forward transitions with a decrease in energy respectively. An example of a similar modification is exhibited in Fig. 6. Such a degeneracy in the energy of some states groups signifies their certain equivalence, i.e. suggests the symmetry addition to the system.

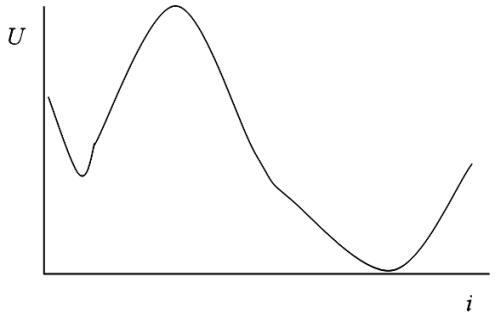

Fig. 6. A modified potential

6. Durability and failure

The ultimate goal of concrete formation is construction of strong and durable material. Thus the main result is not construction of a stronger or more regular structure in itself but of a material structure resistant to failure. A good basis for the kinetic description of composite particles destruction is a kinetic model for cracks growth with an account of their death and reproduction [9]. In the works [10, 11] the correlations of cracks growth mechanics were refined for the case when a crack face forming is of a fractal character.

The classical models for brittle failure is based on the notion of a crack as a linear cut or a cavity with smooth edges. Actually a crack face has fractal properties on an intervening mesoscale of lengths $R_{\min} << R << R_0$.

The lower limit of the fractal model $R_{\min}$ application is correlated to the sizes of a medium granule. The upper limit R_0 is determined by the geometrical sizes of a problem. The size of a crack R is a distance between its edges. As far as a fractal crack is concerned, its length L is correlated with the size R with a power $L = aR^D$, where D is a crack fractal dimensionality. It determines a scale invariance of a set under discussion in given limits. A cascade process of energy transition in a failure process is described with the following chain of equations

$$\Delta U = G_0 \Delta R = N_1 G_1 \Delta R_1 = \ldots = N_n G_n \Delta R_n = \ldots, \quad (16)$$

where we take into the consideration the energy conservation law during a transition from the scale R_n in the form of $N_n G_n \Delta R_n = N_{n+1} G_{n+1} \Delta R_{n+1}$. If we denote $G_n = G(R_n)$, we get the so-called renormalization equation for the function *G*:

$$G(R_n) = \frac{1}{a} \frac{N_{n+1}}{N_n} G\left(\frac{R_n}{a} \right). \quad (17)$$

Let us assume that self-similarity dimensionality is equal to a fractal dimensionality. Then

$$G(R_n) \sim R_n^x, \; x = D - 1. \quad (18)$$

Besides, $G_0 = G(R) \sim R^{D-1}$.

The power parameter $\alpha = 1 - D/2$, i.e. the fractal dimensionality determines the asymptotics of elastic fields stress in the vicinity of a crack tip. If a failure is not ideally brittle, partial energy dissipation will occur during an energy transition from one structural level into another. If the speculation about self-similarity of the energy transition process remains valid, the energy transformation law will take the following form:

$$q N_n G_n \Delta R_n = N_{n+1} G_{n+1} \Delta R_{n+1}, \; q < 1, \quad (19)$$

where $1 - q$ is a proportion of the energy spent in the dissipative processes during a transfer from one structural level into another. In this case, we can obtain $\alpha = (2 - D - \beta)/2$, where $\beta = -\ln q / \ln a$ provides the control of dissipative processes effect.

A unified measure for failure is a number of elementary structural elements that underwent the disruption of the bonds with the neighbouring elements. Microfluctuation take place only at the smallest scale level where scaling properties do not show up. A crack growth itself retains scale invariance properties [12].

Conclusions

The studies carried out are of significant meaning for the understanding of the processes leading to concrete structure formation at the initial stage when physical processes connected with an external action and internal forces dominate over the chemical ones. These processes are most

essential to the stages that follow. The main feature of these processes is their slow character due to the fractal structure and its mechanical inertia on typical time technological scales. While transporting concretes, the blend viscosity depending on the formation of clusters should be considered. This means that during a concrete transfer along the pipes we should take the technological measures into account and prevent coarse grains from penetrating into concrete.

The following methods may be used in order to accelerate the structure formation processes. Metastable granular systems internal stability may be disrupted due to film liquid wetting. This allows to assume that it is possible to construct materials on the basis of liquid wetting self-solidifying polymers with a low viscosity such as that of dry sand.

In order to accelerate the concrete structure formation process, external actions and inner inclusions increasing the degree of the system inner symmetry should be used. The inclusions creating local fields and regular structures at the mesolevel may be effective.

The ultimate concrete strength depends both on its density, strength of the bonds formed and on a structure character. Concrete fractal structure increases power inputs on the formation of cracks as compared to those for the formation of smooth cracks. Besides, the growth rate of fractal cracks is vastly lower than that of smooth ones. This means that controlling the concrete inner structure fractality, i.e. the inner heterogeneity of the relations between its mesoscopic subsystems, we are able to change the strength and durability of the material itself and of the whole construction as well.

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