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Development of Ideas About the Rheological Behaviour of Building Mixtures Taking into Account Fractal-Cluster Processes in Their Structure Formation

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Abstract

Development of theoretical ideas about the mechanism of the rheological behaviour of building mixtures and the experimental assessment of their rheological properties is a relevant area of physiochemical research of materials. To assess the changes in rheological properties when varying the component composition of building mixtures, it is important to use quantitative indicators characterising the microstructure of the mixtures. Revealing the regularities of the formation of heterogeneous microstructures makes it possible to assess their correlation with the rheological properties of building mixtures at the macro level. The aim of the paper is to discuss the results of the implementation of methodological approaches, theoretical modelling, and experimental assessment of the quantitative indicators of the rheological properties of typical building mixtures.

The experimental research methodology is based on the assessment of the rheological properties of heterogeneous dispersed systems (HDS), taking into account fractal-cluster manifestations in their microheterogeneous component. The experiment was carried out using model HDS containing the components of building mixtures. Their rheological properties were determined by rotational viscometry with different compositions of HDS. The fractal dimension *D* was used for a quantitative assessment of the structural and rheological properties and identification of the patterns of their change depending on the composition of mixtures. The value was determined by mathematical modelling.

We analysed model concepts of the rheological behaviour of building mixtures. It was shown that the existing rheological models of an elastic-viscous-plastic medium did not give a complete description of the processes of formation and destruction of the microstructure of concentrated HDS (building mixtures). We carried out an experimental assessment of the effect of the properties of solid phase particles on the change in the structural and rheological characteristics of HDS, taking into account the fractal-cluster principles of their structure formation.

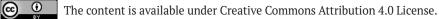
We specified the ideas about the mechanism of rheological behaviour of building mixtures. They take into consideration the processes of the formation and destruction of fractal-cluster formations in the microstructure of HDS. It was shown that the fractal dimension *D* can be one of the quantitative characteristics of the structural and rheological properties. We determined the correlation between the fractal dimension *D* and other experimental rheological characteristics: the ultimate shear stress and effective viscosity. The obtained results can be used to regulate rheological properties and optimise the technological processes for the manufacture of building materials and products.

Keywords: heterogeneous dispersed systems, rheological properties, building mixtures, modelling, fractal-cluster structures.

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1. Introduction

When carrying out physiochemical studies of the structures of building materials, it is important to determine their rheological properties. Rheological properties affect the parameters of technological processes in the construction industry. For example, the rheological properties of mortar and concrete mixtures determine the conditions of the mixing, transportation, moulding, and compaction technologies. They ultimately affect the physical and technical characteristics of finished products and structures [1-21].

Currently, in the production of mortars and concretes, the range and component composition of raw materials with increased dispersity and concentration of solids is expanding. We have multicomponent compositions with fine ground mineral additives, surfactants, organomineral additives, nanosized particles, etc. [2-19]. Constructors apply highly mobile and self-sealing mixtures, whose rheological characteristics ensure preservation of the structure during transportation. At the same time, such mixtures remain highly fluid during the moulding process [2, 3, 9-11]. The mixtures under study are multicomponent, multiphase heterogeneous dispersed-granular systems. Due to the complexity of the study of their structure, we used empirical approaches based on varying the formulation and technology factors and assessing the change in rheological properties without taking into account internal forces and interphase interactions. To study the relationship between the structure and the properties, it is advantageous to implement a methodological approach based on modelling the structure as a multilevel hierarchical system. This approach makes it possible to describe the processes and phenomena that occur at various levels of scale and stages of the structure formation of concrete. During the early stage of coagulation structure formation, on the scale of colloidal and microheterogeneous particles, internal forces are manifested at the micro- and meso-levels of the structure of concrete mixtures (10⁻⁸– 10^{-3} m). In such a system, physiochemical processes of interparticle and interphase interaction are determinant at the interphase boundary and within the "dispersed phase - dispersion medium" phases. The fractal-cluster structures, self-similar at various scale levels, typically form as a result of the manifestation of molecular electrostatic and capillary-film forces [22].

The formation and destruction of aggregated fractal-cluster structures has a significant effect on the manifestation of intra- and interflow interactions, characterised by a change in rheological properties [22]. This predetermines the need for research and design aimed at developing the principles of the rheology of concentrated mixtures. For the systems under consideration, the manifestation of rheological properties depends on the dispersity of solid phase particles, the particle shape and surface roughness, the solids concentration, etc. Revealing the patterns of formation of fractalcluster structures makes it possible to assess the interrelation between structural changes and the dispersion properties. In this case, a quantitative assessment of the structural and rheological properties plays an important role. It is important to take into account fractal-cluster manifestations and the dependence of their change on the properties of solid phase particles.

It is necessary to implement new scientific and practical approaches to analyse self-organising processes, which are difficult to observe directly. We believe that one such approach is the assessment of the rheological properties of building mixtures taking into account the characteristic fractal-cluster manifestations at the microheterogeneous level. The complex application of the theoretical provisions of the rheology of heterogeneous dispersed systems, fractal geometry, statistical physics, and mathematical modelling becomes an effective methodology to expand our knowledge regarding the processes of formation and destruction of the structure of building materials at the micro-level.

Thus, the aim of the work is to implement methodological approaches to modelling and experimental assessment of the rheological properties of multicomponent building mixtures using quantitative indicators that characterise the processes of structure formation, composition, and properties of mixture components. These multicomponent mixtures are mineral pastes containing solid phase particles of various nature.

2. Experimental

As we know, classical models of the rheological behaviour of HDS, including multicomponent building mixtures, have been developed for homogeneous models of the continuum mechanics: the Kelvin, Bingham, and Newton models, etc. [23–25].

The rheological behaviour of the systems under consideration can be described by the following equation [1, 22–25]

$$\tau = \tau_0 + \eta \cdot \varepsilon^n, \tag{1}$$

where τ is the shear stress; τ_0 is the ultimate shear stress; η is the effective viscosity; ε^n is the shear velocity gradient; *n* is the pseudoplasticity index.

We applied experimental methods to model the rheological behaviour of building mixtures. They were based on the determination of the ultimate shear stress τ_0 , the effective viscosity η , or a combination of these values [22, 26]. These integral indicators allow indirect assessments of the viscoelastic characteristics of building mixtures. However, they are not sufficient for a full understanding of how the structure of mixed materials is formed and destructed. When dispersions in the form of pastes, suspensions, foams, and gels are exposed to intense external factors, individual structural elements of mixtures, represented by particles and aggregates of the disperse phase of various concentrations and shapes, undergo changes due to reorientation, disaggregation, etc. (Fig. 1) [25].

For the experimental assessment of the rheological properties of mortar and concrete building mixtures is so complex with regards to the methodology due to the heterogeneity of their structures. This can be explained by the combination of coarsely dispersed components, fine and coarse aggregates, as well as microheterogeneous components: a binder and finely dispersed additives. The manifestation of rheological properties is largely associated with the formation of aggregated fractal-cluster systems in the microstructure of mixtures. For the systems under study, the existing theoretical provisions and models do not provide an exhaustive description of the processes of formation and destruction of the microstructure of concentrated watered dispersed systems and mixtures.

Based on the theoretical provisions [27], the model for the manifestation of the rheological properties of watered concentrated HDS is shown in Fig. 2. The model shows that for the values larger than τ_0 , the formed structure of the dispersion in the form of a percolation fractal cluster breaks down into individual aggregates, which is accompanied by a decrease in the effective viscosity η (Fig. 2a, p. 1 – p. 2). P. 2 – p. 3 zone in Fig. 2a reflects the destruction of interaggregate bonds, the effective viscosity is minimal $\eta_{\text{min}}.$ With an increase in stresses, the bonds break completely, with the formation of smaller clusters and individual particles. This leads to an increase in internal friction and an increase in viscosity (Fig. 2a, p. 3 – p. 4). A subsequent increase in stresses again leads to a decrease in viscosity because of the discontinuity within the system (Fig. 2a, p. 4 – p. 5). Due to the outlined features of the manifestation of properties in the microheterogeneous component, it is important to obtain a quantitative assessment of the structural and rheological characteristics of mixtures.

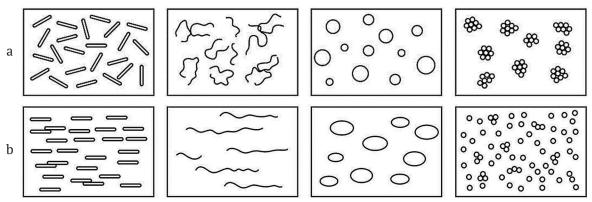


Fig. 1. Schemes of the structure models of dispersed systems without shear (a) and structural changes under shear stress (b)

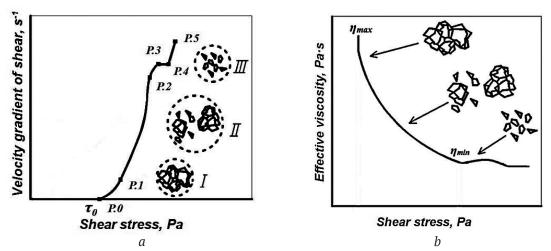


Fig. 2. Models of flow curves of dispersed systems in the form of dependence of the shear velocity gradient (a) and effective viscosity (b) on shear stress

The experiment was carried out on model HDS: mineral pastes containing solid phase particles of various natures, which are used in typical building mixtures. We studied the "cement – water", "sand – water", "limestone – water", and "ash – water" HDS. The dispersity of the solid phase particles was 300, 500, and 700 m²/kg. Rheological properties were determined using rotational viscometry. We determined the correlations of the shear velocity gradient and effective viscosity with the shear stress by varying the composition of the model HDS.

For a quantitative assessment of structural and rheological properties, taking into account fractal-cluster manifestations, we used the function [28, 29]

$$\frac{\eta(\boldsymbol{\varphi},\boldsymbol{\tau})}{\eta_0} = \frac{1 - \boldsymbol{\varphi}_A}{(1 - \boldsymbol{\varphi}_A/\boldsymbol{\varphi}^*)}, \ \boldsymbol{\varphi}_A \approx \boldsymbol{\varphi} \left[1 + \left(\frac{\boldsymbol{\tau}_0}{\boldsymbol{\tau}}\right)^{1/2} \right]^{3-D}, \quad (2)$$

where $\eta(\varphi, \tau)$ is the effective viscosity; η_0 is the liquid phase viscosity; φ_A the effective concentration of clusters; τ_0 is the ultimate shear stress; τ is the shear stress; φ is the solid phase concentration; φ^* is the critical concentration of clusters; *D* is the fractal dimension.

The forming structures were assessed based on the value of the fractal dimension *D*, which characterises the self-similarity properties of inhomogeneous, disordered objects and systems [22]. In this paper, the value of *D* was calculated by numerical modelling in the mathematical program Maple [29]. The modelling required solving mathematical equation (2) using the experimental data:

$$\eta = f(\tau), \tag{3}$$

where η is the effective viscosity; τ is the shear stress.

In the process of modelling we built a dependence diagram (Fig. 3). It corresponded to the experimental curve, when equation (2) was solved adequately.

We applied the method of mathematical modelling with the calculation of the fractal dimension *D* in order to develop ideas about the processes of formation of aggregated fractal structures of watered dispersions. By studying the values of *D*, we can quantitatively characterise the effect of the properties of solid phase particles on the structural and rheological parameters of dispersed systems.

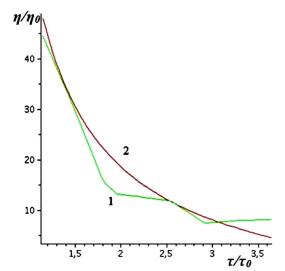


Fig. 3. Fragment of building rheological curves in the Maple program: 1 – experimental curve; 2 – the curve built by solving the mathematical equation

Optical studies of the structure of watered dispersed systems were carried out using a Biolam D-12 (Lomo, Russia), combined with a digital camera (computer-controlled Olympus SP-500 UZ). A red LED was used as a light source for the microscope. The equipment made it possible to obtain an image with a resolution of up to 300 pixels/inch with a clear outline at 10x magnification. The fractal dimension *D* was determined using the aforementioned mathematical model in the Maple program [29].

3. Results and discussion

The experimental results of the study are consistent with the known theoretical concepts of the mechanism of rheological behaviour of concentrated HDS (Fig. 4). The areas with a minimum viscosity (Fig.4a, p. 2 – p. 3) were revealed, an increase in viscosity was observed at high shear stresses p. 3 – p. 4. Then, there was again a decrease in viscosity with the discontinuity of the system at p. 4 – p. 5. From a practical point of view, it is important to note that sometimes the early manifestation of an area with

an increase in viscosity (Fig. 4a, p. 3 - p. 4) at high shear stresses during technological processes does not allow the effectuation of an area that reflects the maximum dilution of mixtures.

The noted features of the manifestation of the rheological properties of the studied dispersions result from the hydrodynamic properties of particles. They are caused by the processes of the formation and destruction of the aggregated fractal-cluster structure. This explanation is confirmed by the results of optical studies of the structure of the dispersed system (Fig. 5).

In the course of the analysis and generalisation of experimental data, the impact of the properties of solid phase particles on the change in the structural and rheological characteristics of dispersed systems was estimated. The obtained data (Fig. 4, Table 1) shows that the type and nature of the solid phase particles have a significant effect on the main rheological properties of HDS. The ultimate shear stress τ_0 varies from 26 Pa (for the "limestone-water" system) to 466 Pa (for the "ash-water" system).

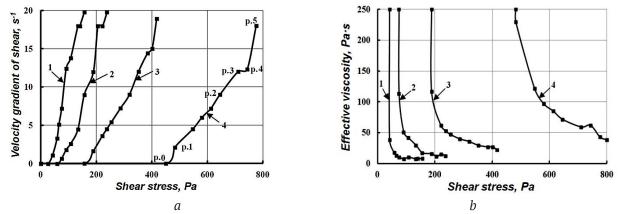


Fig. 4. Experimental rheological curves of dispersed systems in the form of dependence of the shear velocity gradient (a) and effective viscosity (b) on shear stress (water/solid ratio is 0.32, dispersity is 500 m²/kg)

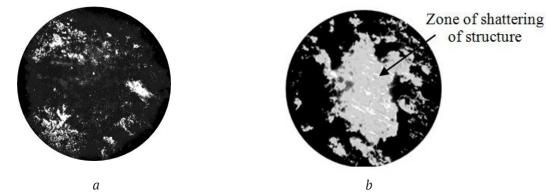


Fig. 5. A change in the structure of the "sand-water" dispersed system with shear stress under ×10 magnification (water/solid ratio is 0.32, dispersity is 500 m²/kg): a) the system without shear; b) the system after shear

The minimum effective viscosity η_{min} for these systems varies from 7 Pa·s to 38 Pa·s.

We determined the dependence between the values of the fractal dimension D and the experimental integral characteristics, which are the ultimate shear stress τ_0 and effective viscosity η_{min} (Table 1). It was shown that the fractal dimension D decreases in the studied systems when τ_0 and η_{min} increase. It can be assumed that the observed correlations are due to the properties of the solid phase particles. In studies [28–30], it was shown that ash particles have a more developed and rough outer and inner surface, as compared to the other studied particles. This contributes to the formation of a dispersed system with a more branched fractal-cluster structure, where the manifestation of internal forces provides the highest values of $\tau_{_0}$ and $\eta_{_{min}}$ with the minimum fractal dimension D = 2.31.

We assessed the effect of the dispersion of solid phase particles on the change in the structural and rheological properties using the example of the "sand – water" system, with a constant water/ solid ratio W/S = 0.28 (Fig. 6, Table 2).

As expected, with an increase in the values of dispersity, an increase in τ_0 and η_{min} is characteristic. If we increase the dispersity of the studied system from 300 to 700 m²/kg, τ_0 increases from 226 Pa to 389 Pa, and η_{min} increases from 21 Pa·s to 31 Pa·s. These observations can be explained by the manifestation of internal forces and a change in the nature of the forming fractal-cluster structures. This change results in a decrease in the fractal dimension *D*. Thus, the value of *D* shows that with an increase in dispersity in the watery system, a more branched microstructure is formed, with a large number of inter-particle and inter-aggregate contacts. It is

Table 1. Influence of the type of dispersed systems on their structural and rheological characteristics (water/solid ratio of 0.32, dispersity of 500 m^2/kg)

Indicator	Dispersed system			
	"limestone-water"	"sand-water"	"cement–water"	"ash–water"
τ ₀ , Pa	26	58	156	466
η _{min} , Pa∙s	7	11	22	38
D	2.64	2.61	2.57	2.31

Table 2. Influence of the dispersity of solid phase particles on the structural and rheological characteristics of the "sand-water" system (with a water/solid ratio of 0.28)

Потгородоти	Dispersity, m²/kg			
Показатель	300	500	700	
τ ₀ , Pa	226	258	389	
η_{min} , Pa·s	21	23	31	
D	2.7	2.65	2.45	

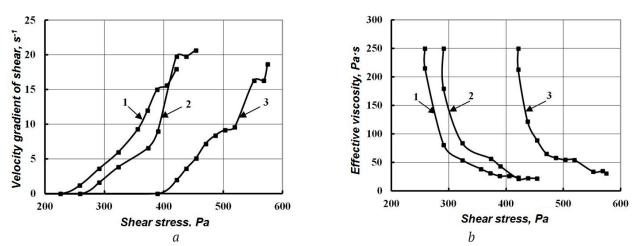


Fig. 6. Experimental rheological curves of the "sand-water" dispersed system at water/solid ratio of 0.28 in the form of dependence of the shear velocity gradient (a) and effective viscosity (b) on shear stress

characterised by a higher void content, while the fractal dimension *D* decreases from 2.7 to 2.45.

4. Conclusions

The obtained research results expand the understanding of the mechanism of rheological behaviour of building mixtures, demonstrate the formation and destruction of fractal-cluster formations in the HDS microstructure. It was shown that the fractal dimension D can be used as a quantitative characteristic of the processes of structure formation of HDS at the micro level, as well as for additional assessment of their structural and rheological properties. This value can be determined by mathematical modelling using specialised software. We determined the interrelation between the fractal dimension D and such experimental rheological characteristics as the ultimate shear stress τ_0 and effective viscosity η . These parameters vary depending on the properties of the solid phase particles of the HDS components, which are the basis of typical building mixtures. The obtained results can be the basis for regulating the rheological properties of building mixtures and optimising the technological processes of mixing, transportation, and moulding.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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