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BUILDING COMPOSITES AS OBJECTS OF SYSTEM ANALYSIS

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Abstract. The article is devoted to the consideration of the material science problems from the standpoint of the general systems theory. The method of dialectical pairs "part-whole", "continuous-discrete" and "randomness-necessity" is used, corresponding to the general system coordinates, on the basis of which the third emergent component of a synthetic nature arises. System-theoretic methods make it possible to concretize model representations using elements of the crystallization theory of dynamic information theory. System representations are used to refine the concept of an interface in materials and heterogeneous systems. The dialectical unity of atomistic and geometric concepts in the theory of the formation of a new phase in binders is shown. The probabilistic and informational description is considered as a realization of the dialectical pair "randomness-necessity". The concept of information is closely related to choosing one or more options from many and remembering the choice made. The choice can be made as a result of the action of external forces (information reception) or due to the unstable behavior of the system (the emergence of new information). The process of structure formation can be considered as the dynamics of an information system. The dynamics of the information system should include interactions that provide any structure-forming particle with the possibility of transition between the areas of influence of stable states with subsequent attraction to one of them. The formation of structures in a binder from the position of dynamic information theory is considered as the reception of information. It is necessary to perform work, due to which the system will go into one of the stable states for the reception of information. The formation of new elements and structures – pores, capillaries and cracks is considered as a transition of the system to its own potential minima – the epigenetic landscape of the material as an emerging system. In this case, part of the energy will be lost in the process of dissipation, and the physical entropy will increase by a value exceeding the amount of information received. Such an effect can be considered as the "price" of the observed macroscopic process of self-organization. The involvement of systems theory also helps, on the basis of the principle of adequacy, to indicate the way for choosing a model object that is optimal for studying the processes of structural transformations of a material.

Keywords: composite building materials, dispersed systems, system approach, structure formation, dialectical pairs.

Introduction. Composite building materials (CBM) are presented as open complex non-equilibrium systems in modern materials science [1]. The properties of such materials, including structural-mechanical ones, can be described by several methods. In the first category of methods, spatial averaging techniques are applied (averaging over structural components) and a transition is made to the reduced characteristics, such as the modulus of elasticity or shear. In the second category of research methods, a significant departure from the concept of a material as a continuous medium is made, which corresponds to the results of modern studies of structural-mechanical and

physical-chemical properties [1]. Since it is rather difficult to find among a set of mathematical models and representations strictly suitable for reflecting new facts based on a structure-oriented approach, many mathematical models of various types, both continuous and discrete, are used in this area [1-4]. For the same reason, when studying the behavior of such systems, a verbal description is often used [1, 5].

Analysis of recent research. To describe the properties of systems of the type under consideration, it can be considered productive to apply the principles of the self-organization theory, as well as the general systems theory [1]. The wide possibilities of these methods can be applied to specific categories of materials. In particular, the multivariate behavior and properties of CBM, for example, mineral or polymeric materials, are in good agreement with the wide possibilities of the methods under consideration. This work is devoted to the analysis of the possibilities of applying the corresponding methods to composite materials of the two types considered.

One view of general systems theory is that it is a more concrete embodiment of the dialectic [5]. In systems theory, any object can be represented in a kind of coordinate space, each axis of which corresponds to a dialectic pair (DP). The considered system or approach in this case is a compromise, and sometimes a non-trivial synthesis, and complements pairs to triads. The dialectical theory of systems is, by necessity, explicitly or implicitly, ternary, triadic. Some dialectical pairs and their dialectical synthesis seem to be especially important for the problems of materials science.

Targets and goals. The purpose of this work is to increase the interpretative power and predictive ability of traditional physical and chemical methods of building materials science by using together with them methods based on the general systems theory, and to apply the built version of the system approach for the theoretical study of mineral and polymer composite materials. The corresponding tasks are, according to the applied method, the selection of the most important dialectic pairs and the disclosure of their content using the statistical theory of solidification of composite materials and dynamic information theory.

Objects and methods of research. The objects of research are the composite materials based on mineral (gypsum) and polymeric binders, considered as system**s.** In this case, a system is understood as a secondary integrity that arises when primary, relatively integral objects are combined, accompanied by the emergence of new properties in this association that are not reducible to the original ones. Methods for studying objects of materials science as systems are hierarchical. The upper, structural-logical level, is associated with the use of dialectical system methodology. This abstract level is concretized with the help of probabilistic methods of solidification theory and information theory, which allow one to proceed to specific recommendations on the choice of objects and methods of experimental research based on the adequacy principle.

Research results. The analysis of structure formation processes in this work is carried out in accordance with the following algorithm. At the first stage, with the help of system-theoretical representations, a set of dialectical pairs is distinguished, each of which is represented by one of the "coordinate axes" that allow classifying materials and the processes occurring in them. It is determined what is a dialectical synthesis for each direction. Then, the considered structural-logical approaches are concretized using a number of physical-chemical methods. In accordance with this plan, the following dialectic pairs (DP) are considered.

DP "part-whole". Such a pair is determined by a compromise in establishing the boundary between the system and the environment, i.e. by specifying a surface bounding the system. This surface also provides all kinds of interactions with the environment. Thus, a mineral or polymer binder, taking into account its intended purpose (functioning in a structure), can be considered as an open hierarchical system with its own scale levels, at each of which the issue of the interface has its own special aspects. At the micro level, a binder material, for example, gypsum stone, is a polycrystalline body, which also includes a finely crystalline component [6], in which irregularly shaped crystals are linked into a network crystallization structure. The resulting structure often has the character of a percolation cluster [3], and its boundaries coincide with the surfaces of the sample under study. The description of the structure of other materials, for example, polymer compositions, is qualitatively similar at the level of cluster structural units [7]. One of the reasons for the qualitative similarity is the

systemic separation of structural levels, for example, the separation of the macroscopic level from the molecular level. In this case, a qualitatively similar behavior at the macro level arises despite the significantly different molecular structure of the materials. In the study of such structures, the problem of the interface is solved by constructing a cellular partition of a three-dimensional space and calculating cellular characteristics that turn into dimensional ones for extremely small cell sizes. In this case, the surface, linear, and point features included in the system are naturally taken into account. From a macroscopic point of view, the material should be considered in operational conditions – as a structural element [1] or by fixing the boundary conditions at the interface of the corresponding blocks.

The problem of the existence of an interface can also, in turn, be representable by a DP. On the one hand, the classical model representation is the model of the interface as a smooth surface, the intersection of which is accompanied by an abrupt change in the properties of the corresponding phases. On the other hand, the atomic and molecular structure of the corresponding composites makes it possible to represent the material in the form of a continuous network structure, and the formation of individual structural blocks (clusters) is associated with an increase or a maximum decrease in the number of bonds in a molecular or polycrystalline network (Fig. 1). With such a structure, it is difficult to speak of an interface as such.

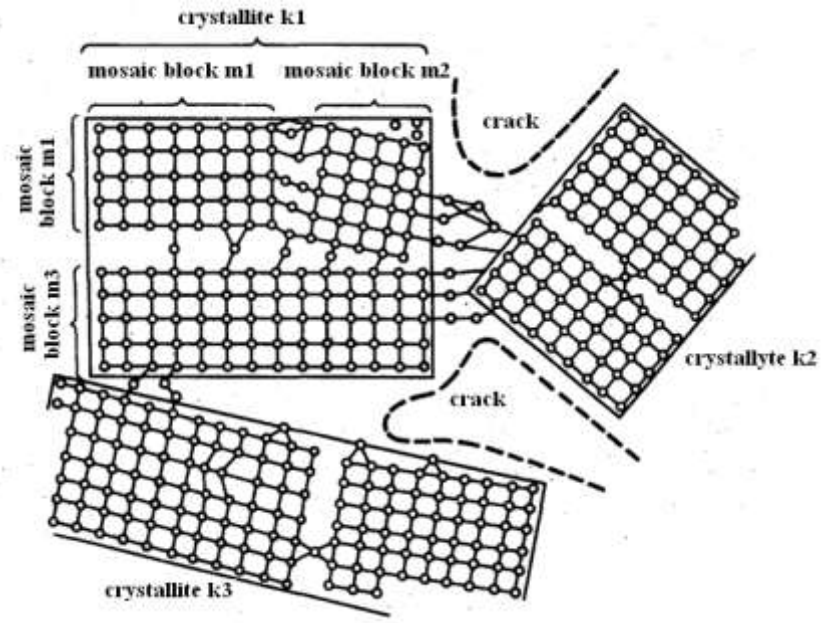

Fig.1. Representation of the material as a mesh polystructure

DP "continuous-discrete". For various reasons, a universal feature manifests itself - a tendency to grouping in space with the formation of structures, as well as in time, with the formation of consistent tempo-rhythms and being synchronized, for all material formations from molecules to organisms. The space-time structures formed in materials have the ability to interact with each other to form a superstructure. Particles of material, forming hierarchical fractal-like systems in space, "filling" the space and modeling a continuous medium, are characterized by both discreteness and continuity. This dialectical pair is one of the most essential for material systems. For different spatial scales, the mechanisms of formation of structures are different, but here, too, similarity considerations remain valid. Another version of theories going from continuous to discrete is the representation of discrete objects as spatially separated maxima of the concentration or distribution function [2].

To study the process of hydration, which occurs simultaneously with structure formation in a binder dough, for example, based on gypsum, the state of the near-surface layers of crystals of the initial binder $CuSO_4 \cdot 0.5H_2O$ is extremely important. This is one of the factors determining the further course of the process – the predominance of topochemical processes of hydration, as a result of which the formation of a finely crystalline product is likely, or the path associated with the dissolution of the binder, the transfer of ions through a layer of saturated solution, nucleation and

recrystallization according to Le Chatelier-Rehbinder [4]. Here, as in the process of interaction of a binder with surface-active additives, the molecular level of structure organization is important. The processes occurring at the interfaces of different scale levels are decisive in the formation of the structure of polymer composites [8, 9].

The next level of structure-forming processes in terms of scale is directly related to the formation of a new phase nuclei from a supersaturated solution and their growth [10]. Most mathematical models do not take into account the structure of nuclei and consider the evolution of the distribution of nuclei by the number of atoms included in them *n*, *n>>1*. At the same time, at each stage, the attachment of new atoms to the nucleus is associated with the fluctuation passage of a large number of intermediate states – "active complexes", each of which corresponds to different free energy maxima *ΔF*. Consideration of structures of this type is carried out using the theory of Markov processes, taking into account the possibility of transition to the nearest intermediate state. Kolmogorov's equations are an adequate mathematical apparatus. For the number of nuclei *Z(n,t)* containing *n* atoms by time *t* we have (1):

$$\frac{\partial Z(n,t)}{\partial t} = -Z(n,t)\left[P_+(n)+P_-(n)\right]+Z(n-1,t)P_+(n-1)+Z(n+1,t)P_-(n+1), \quad (1)$$

here $P_+(n)$ and $P_+(n)$ are the probabilities of addition and detachment of an atom for a nucleus with size *n*.

Neglecting in (1) terms above the second order of smallness allows us to proceed to the diffusion process and the Fokker-Planck equation (2):

$$\frac{\partial Z(n,t)}{\partial t} = \frac{1}{2}\frac{\partial^2}{\partial n^2}\left\{\left[P_-(n)+P_+(n)\right]Z(n,t)\right\}+\frac{\partial}{\partial n}\left\{\left[P_-(n)-P_+(n)\right]Z(n,t)\right\}. \quad (2)$$

If we assume that the intermediate states are in equilibrium with the solution (a small number of nuclei formed), the principle of detailed equilibrium (3) arises:

$$\begin{aligned} b(n)P_+(n) &= b(n+1)P_-(n+1) \\ b(n)P_-(n) &= b(n+1)P_+(n+1) \end{aligned}, \quad (3)$$

here *b(n)* – is the equilibrium distribution function (4):

$$b(n) = N\exp\left\{-\frac{\Delta F(n)}{kT}\right\}. \quad (4)$$

From (1) and (3) in approximation (5):

$$\frac{\partial P_\pm(n)}{\partial n} \langle\langle P_\pm(n), \quad (5)$$

we can derive the Zeldovich equation (6) [10]:

$$\frac{\partial Z(n,t)}{\partial t} = \frac{\partial}{\partial n}\left[P_+(n)\,b(n)\frac{\partial}{\partial n}\left(\frac{Z(n,t)}{b(n)}\right)\right]. \quad (6)$$

The transition state method for the probabilities of adding and removing an atom $P_\pm(n)$ gives approximation (7):

$$P_\pm(n) = \tilde{n}\,\omega\exp\left\{-\frac{U}{kT}\mp\frac{1}{2kT}\frac{d\Delta F}{dn}\right\}, \quad (7)$$

where $\tilde{n}$ – is the number of atoms near the surface of the nucleus, ω – is the oscillation frequency near the surface of the nucleus, *U* – is the average height of the activation maximum.

The critical nucleation concentration (CCN), which is traditional for the macroscopic description of formation and growth, enters the stochastic description through the quantities $\tilde{n}$ and $P_\pm(n)$ from (7).

At a certain size n^*, the free energy reaches a maximum (8), which corresponds to the critical size of the nucleus:

$$\left.\frac{d\Delta F(n)}{dn}\right|_{n^*} = 0. \quad (8)$$

Since the activation energy is U>>kT, then almost all the nuclei that have reached $n=n^*$ will be realized in the centers of the new phase. Due to the same relation, each such nucleus can be considered as an attracting center, a spatial attractor for ions.

The considered classical model of nucleation, being an essential idealization, does not answer many questions of the structure formation theory. In particular, the probability of joining new structural units to existing ones depends on the nature of the interface and, in a discrete representation, on the structure of the nearest environment of the structural units under consideration. Such a quantity is not isotropic (it will differ in directions). Despite the simplification, the above representation indicates the main trends – the stochastic nature of the formation of nuclei of a new phase and the existence of its critical size.

One of the essential heuristic analogies, which is valid for disperse systems, is the "atom – colloidal particle" analogy. It allows transferring, with appropriate correction, approaches and methods of description between structural levels of different scales. In particular, at the next stage of growth, the formation of groups of particles – clusters and groups of clusters, and the formation of a hierarchical structure of the material [2] is observed. It is fundamentally different from the structure of crystalline bodies, the ordering here is of a probabilistic-statistical nature. The model of close packing and a constant coordination number of a cluster-forming particle seems to be rarely implemented due to the uneven distribution of the free energy of the particle surface. Clusters and their groups form a poorly ordered system, more characteristic of a liquid (short range order), hence the need for a statistical description. The approach presented in [1, 2] is generally valid for their formation. It is important to take into account that the emerging clusters, like the emerging nuclei for ions, are spatial "traps" that attract sets for colloidal particles.

The formation of a coagulation structure is associated with the interaction of colloidal particles. It is carried out due to a set of forces of various nature – dispersion, dipole-dipole, electrostatic (associated with the mutual influence of double layers) interaction due to the structural-mechanical barrier formed, in particular, by the surface layers of adsorbed particles. As a result, the representation in the form of a sum (9):

$$U(r)=\sum_i U_i(r)=U_{disp}(r)+U_{electrost}(r)+U_{str-mech}(r)+\cdots, \quad (9)$$

provides an adequate description of the complex nature of the interaction of colloidal particles.

One of the model approximations can be a refusal to consider specific types of contributions to potential energy or simplified assumptions about them. In the simplest case, it is possible to take into account the only qualitative feature and replace the considered potential (9) with a model one – a "comb" or a sum of sinusoids. The "comb" method turns out to be useful, in particular, in studying the spatial structure of [11] proteins. The dynamics of particle interaction during coagulation structure formation can then be considered by the Kramers or Langevin method [12].

Coagulation and crystallization clusters at the next stages form a percolation hierarchical structure, due to which the system undergoes setting and eventually forms a stone-like body due to the enclosing nature of the percolation cluster of interparticle contacts of various nature (coagulation and crystallization contacts, interparticle adhesion contacts). In addition, it should be noted the implementation of deep minima in effective potentials (9), due to the existence of the "material" connections discussed above. Thus, the interaction and mutual transition between discrete (spatial structures) and continuous (distribution of binder particles) is characteristic of all stages of structure formation.

DP "randomness-necessity". In any disperse system, there are interactions that can be fairly conventionally divided into deterministic and stochastic, and their further study can be carried out by various methods, including combined ones. In a number of cases, such a division is due to physical reasons, for example, a significant difference in the masses of the molecules of the solution and the Brownian particle. The probabilistic description is also justified in the case of unstable chaotic dynamic regimes. For the corresponding systems (of a deterministic nature!) it becomes possible to indicate equations similar to diffusion ones. Often the selection of the stochastic part is determined subjectively by the degree of difficulty of the study. Randomness should be recognized

as an unknown necessity, and necessity as a common mechanism for generating randomness.

All these mutual transitions can be observed on the example of structure formation in materials. One of the most productive ways of considering this DP is based on information theory.

The concept of information involves choosing one or more options out of many and remembering the choice made. The choice can be made as a result of the action of external forces (reception of information) or due to the unstable behavior of the system (the emergence of new information). The process of formation of structures can be considered as the dynamics of the information system. It should include interactions that provide any structure-forming particle with the possibility of transition between the regions of influence of N stable states with subsequent attraction to one of them. Then the maximum information (information capacity) that the system can prepare is (10):

$$I_{max} = log_2 N . \quad (10)$$

On the other hand, there are particles in the system for which it is possible to determine the thermodynamic probability *W*, including the values of their coordinates and momenta. The definition of physical entropy in bits through W also corresponds to micro information I^{mic} (11) [13]:

$$I_{max}^{mic} = \tilde{S}_{max} = log_2 W . \quad (11)$$

The Brilouin principle of equivalence of information and physical entropy (12) is fulfilled for micro information:

$$I^{mic} + \tilde{S} = I_{max}^{mic} = \tilde{S}_{max} = log_2 W . \quad (12)$$

The number of stable states *N* is usually much less than *W*. For the reception of information (10), it is necessary to perform work, due to which the system will passes into one of the stable states. In this case, part of the energy will be lost in the process of dissipation, and the physical entropy $\tilde{S}$ will increase by a value exceeding the amount of information received. It is fair to consider such an effect as the "price" of the observed macroscopic process of self-organization. Due to dissipative properties, there is a transition to stable states, the lifetime of which τ is determined by the height of the potential barrier between them (13):

$$\tau = \frac{h}{kT} exp \frac{U}{kT} . \quad (13)$$

Let us pay attention to the fact that in the process of structure formation, the nuclei of a new phase and growing clusters ensure the existence of spatially stable states, which can be estimated by the sum (14):

$$N(t) = \sum_{n^*}^{n^{max}} Z(n,t) . \quad (14)$$

It should be noted that (14) refers to the supercritical nuclei of a new phase – growing microcrystals.

A rough estimate also gives (15):

$$N_{max} = \frac{N_{am}}{n^*} , \quad (15)$$

here N_{at} is the number of mobile atoms involved in the growth of the nucleus. Such an approximation is possible, since starting from $n=n^*$ the process can be considered irreversible, $U>kT$.

Thus, the process of formation of the embryonic structure and the further stage of growth can be interpreted as a kind of reception of information, the transformation of a part of micro information (11, 12) into (macro) information (10). In this case, due to the dissipation of the interaction energy of particles and the growing structure, the physical entropy increases, and the centers of the new phase correspond to stable states. A similar conclusion applies to many phase transitions accompanied by crystallization. During the structure formation of cluster systems, colloidal particles interacting with the total potential (9) are considered as "atoms", and a growing cluster can also be recognized as a kind of spatial stable state.

In the process of structure formation for materials of various nature, the formation of new elements and structures is observed – pores, capillaries, cracks. Even more complex structures are formed in composite materials with the participation of filler particles. The approach considered above can be extended to complex structures, if we take into account the possibility of complicating the nature of the potentials in the corresponding systems and the emergence of multi-stability. The geometric shape and other parameters of the emerging structures correspond to their own minima of the potential – the epigenetic landscape of the emerging system. General characteristics of a complex picture of structure formation with the emergence of new elements and simplified representations associated with the theory of nucleation are the formation of macro information (associated with the type of the emerging structure and the corresponding minima of the potential function – attractors) and the increase in physical entropy due to this according to (12).

The transition of a part of micro information into macro information is also carried out in the processes of destruction of the material. The microscopic level here should be recognized as the structure of the phase boundary (inter-cluster interfaces, pores and technological cracks) or the corresponding network structure. The formation of fracture cracks as a result of the imposition of internal and external deformations leads to the implementation of a stable mutual arrangement of spatial units (crystallites, atoms and molecules at the molecular level), corresponding to the destroyed material of the sample or structure (11, 12).

For several scale levels, it is possible to observe patterns of interaction and mutual transformation that are characteristic of this DP.

Conclusions. There are other DPs that play the role of coordinate axes in the study of systems. Among them, we note the following: "statics-dynamics", "inertia variability". All of these have been implicitly affected by the consideration of the above DPs. Conclusions. Thus, the analysis allows us to assume that the principle of adequacy should be recognized as a criterion for choosing a model approach to studying the mechanisms of structure formation: for each of the considered system coordinates, the complexity of the object must correspond to the covered complexity of the study method, and the target setting is an additional limitation. The considered "axes" are of the greatest interest for us. It seems desirable to reduce the complexity in other directions for the experimental study of structure formation. In particular, the selection of the binder material can be carried out in such a way as to reduce the complexity and variety of purely chemical processes (hydration). Such materials are mono-mineral binders. One of the possible options for the model system for the direction under consideration is gypsum binders and, in particular, gypsum $CaSO_4 \cdot 0.5H_2O$ directly, the other is polymeric materials, for example, epoxy resin. The processes of setting and hardening of such materials go through various stages of structure formation, which can be described by the proposed methods of systems theory.

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БУДІВЕЛЬНІ КОМПОЗИТИ ЯК ОБ'ЄКТИ СИСТЕМНОГО АНАЛІЗУ

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Анотація. Стаття присвячена розгляду завдань матеріалознавства з позиції загальної теорії систем. Використано метод діалектичних пар «частина-ціле», «безперервне-дискретне» та «випадковість-необхідність», що відповідають загальносистемним координатам, на основі яких виникає третя емерджентна складова синтетичного характеру. Теоретико-системні методи дозволяють конкретизувати модельні уявлення за допомогою елементів теорії кристалізації та динамічної теорії інформації. Системні уявлення застосовуються для уточнення поняття границі розділу у матеріалах і гетерогенних системах. Показано діалектичну єдність атомістичних та геометричних уявлень у теорії утворення нової фази у в'яжучих матеріалах. Імовірнісний та інформаційний опис розглядається як реалізація діалектичної пари "випадковість-необхідність". Поняття інформації тісно пов'язане з вибором одного або кількох варіантів багатьох і запам'ятовування зробленого вибору. Вибір може здійснюватися внаслідок дії сторонніх сил (рецепція інформації) або через нестійку поведінку системи (виникнення нової інформації). Процес утворення структур можна розглядати як динаміку інформаційної системи. Вона повинна включати взаємодії, що забезпечують будь-якій структуроутворюючій частинці можливість переходу між областями впливу стійких станів з наступним тяжінням до одного з них. Формування структур у в'яжучому матеріалі з позиції динамічної теорії інформації сприймається як рецепція інформації. Для рецепції інформації необхідно зробити роботу, за рахунок якої система перейде в один із стійких станів. Формування нових елементів і структур – пор, капілярів і тріщин розглядається як перехід системи до своїх власних мінімумів потенціалу – епігенетичного ландшафту матеріалу як системи, що формується. При цьому частина енергії загубиться у процесі дисипації, а фізична ентропія збільшиться на величину, що перевищує кількість отриманої інформації. Такий ефект справедливо розглядати як «ціну» макроскопічного процесу самоорганізації, що спостерігається. Залучення теорії систем допомагає також з урахуванням принципу адекватності вказати шлях вибору модельного об'єкта, оптимального для дослідження процесів структурних трансформацій матеріалу.

Ключові слова: композиційні будівельні матеріали, дисперсні системи, системний підхід, структуроутворення, діалектичні пари.

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