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Terminology and definition of the amorphous substances

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Abstract: It is attempted to create a clear physical and mathematical basis of the definitions of basic and most general notions in the field of studying the structure of the amorphous materials.

Keywords: *amorphous materials, disordered systems.*

Today one may state quite unhesitatingly that any substance could be produced in the amorphous state. The only question is in what technological conditions the particular substance could be produced in the particular state and in what temperature and time conditions it could exist, i.e. be stable. Such approach to the problems of the amorphous state transfers the emphasis of the studies from the parameters of the substance ability of amorphization to the problem of clarifying their general nature and the ways of obtaining the domain of the parameters and criteria of their stability. In this case, it is particularly important to clarify the general specific character of the atomic structure of the amorphous substances.

The fact that the terminology used in studying the amorphous substances is not strict and, in many cases, ambiguous and 'vague', does not favor the studies of unordered substances. Different researchers treat a number of terms differently. As consequence, disputable questions arise frequently not in fact of the obtained results but they are of purely terminological character. Therefore, in this paper we attempt to create a clear physical and mathematical basis of the definitions of basic and most general notions in the field of studying the structure of the amorphous materials.

No rigorous, internally consistent system of principal definitions and terms in the field of studies of the amorphous substances, which takes into account modern knowledge and is acceptable for overwhelming majority of specialists, is available now [1–3]. Partly, the reasons for this are related to the insufficient study of a series of the most important problems pertaining to the nature of the amorphous substances. According to comment [4], in most cases the principal negative moment concerns a faulty approach to the analysis of the general nature of these objects. First, this is one-sidedness of investigation, when all attention is concentrated on the analysis of certain processes with complete ignorance of other ones. Second, differentiation of definitions of internal and external categories is insufficiently exact. For example, the amorphous substance properties, which result from the use of certain technology, are treated as the manifestation of its internal essence. Third, the notions of 'general' and 'particular' are often confused. And fourth, the most of the leading specialists in this field underestimate the problem of terminology. Therefore, now there exist a great number of terminological confusions in studying the amorphous substances.

At the same time, the activity in the area of studying the amorphous substances is being growing. New amorphous materials appear, more and more specialists are interested in the problems of the disordered systems, new models, ideas and approaches are being suggested. In such conditions, the problem of construction of a consistent terminological system in this field becomes more and more topical. The terminological problems themselves are not crucial, but terminological clarity and unambiguity are an important condition of prevention of the losses and serious distortion of information during its transmission from one group of researchers to another one, which deal with the same problem, but study them from the another standpoint. To ensure effective mutual communication and understanding of different research groups there exists a necessity to establish a generally accepted 'language' in this field. It is extremely desirable that the terms of the above 'language' will more completely reflect the real properties of the amorphous substances. The main problem in creating such 'language' is related to the lack of commonly accepted qualitative and, especially, quantitative criteria, which can define the framework of the use of any term [2, 5].

There is a wide spectrum of opinions concerning the definition of the amorphous substances. Both organic and inorganic liquids are attributed to the amorphous materials [6]. They are identified as non-crystalline materials and in case of their definition their types are simply specified: liquid metals and semiconductors, glasses and evaporated or alternatively deposited amorphous films belong to the non-crystalline substances [7]. More cardinal is the viewpoint of the author of Ref. [1]. He states that the amorphous materials are not as unique and solitary as the crystals, which are limited by a few crystallographic forms. The amorphous materials incorporate a wider but less specific circle of objects of different nature, thus, they have no exact unified definition. Other researchers [8, 9], in turn, note that it is easier to say what the amorphous materials are not, than state, what they are.

When defining the amorphous materials, most of the scientists emphasize on the particular specific properties or groups of properties inherent or absent in them. In particular, the amorphous substances include those having macroisotropic properties, producing arbitrary shaped surfaces at breaking or cleaving: most frequently – the conchoidal cleavage, lack of the crystalline regions both in the compact and disperse state [10]. In Ref. [11], non-crystalline objects below the vitrification temperature are considered the kinetically frozen thermodynamical systems. The author of Ref. [12] defines the amorphous substances as the solid state, in

which, in given conditions of application of shear strain to the sample, the manifestation of irreversible viscous flow could be neglected. In this relation, the amorphous substances may be considered the super-cooled liquid with large viscosity factor $\eta \geq 10^{19}$ P [13]. The lack of the melting point is also frequently considered as the attribute of the amorphous materials [14]. Specific and, at the same time, terminologically vague definition of metallic glass is given in [15]: this is the amorphous solid having two or more chemical elements with different cluster sizes and/or different cluster formation temperatures.

A large part of researchers takes the peculiarities of the amorphous substances structure as the basis for their definition. Such approach has quite rigoro-us mathematical substantiation, because the diffuse smeared X-ray, electron or neutron diffraction patterns are the main characteristic experimental attribute of the amorphous materials. They reveal several smeared maxima with specific large half-width and drastic decrease of intensity $I(s)$ with diffracted radiation scattering angle (or scattering vector module s) (fig. 1). These patterns reflect the structural peculiarities of the amorphous substances. In this case the similar diffraction patterns arise for the different amorphous objects, which may differ from each other by their specific structure. Such situation is completely similar to that in crystals, where different clystallographic forms have the same type of diffractograms in a form of a system of single sharp discrete point reflexes (fig. 2).

Below the structure of different substances will be the principal object of our analysis. In such studies, one has to point out clearly two qualitatively different structural levels.

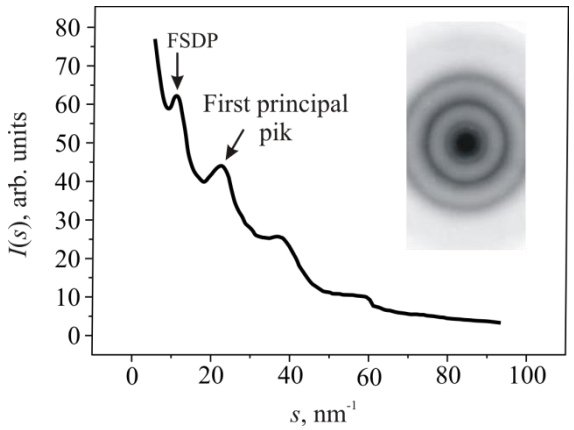

Fig 1. Typical diffractogram and electronogram (inset) of the amorphous substances (in this case – the amorphous $As_{40}Se_{60}$ film)

One of these levels specifies the character of the spatial location of the structural particles of the substance stipulated by their shape, character and intensity of interaction forces. In structural studies, the visual presentation of such structure is often used. To do this, it is assumed that in any condensed state phase of given substance for the most elements of structure one may find the correspondence between the positions of certain structural particles and the nodes of certain spatial lattice. For crystals this allows one to obtain the spatial periodic crystalline lattices. According to this, graphical images in a form of unordered spatial carcasses or networks with nodes corresponding to spatial locations of atoms are used in studying the structure of the amorphous substances as well. Therefore, the majority of researchers call this level the atomic network (carcass) structure. On this level the main elements of the structural ordering in the amorphous substances are revealed: the short-range order (SRO), the intermediate-range order (IRO) and the mesoscopic ordering. All the above elements of the atomic network structure in the amorphous substances make their specific contribution into the diffraction data (fig. 1). Note also that in different substances the separate atoms, ions, molecules, macromolecules or other atomic formations are chosen as the structural particles. In general, the structural particles of the different scale levels can be used to describe the particular substance.

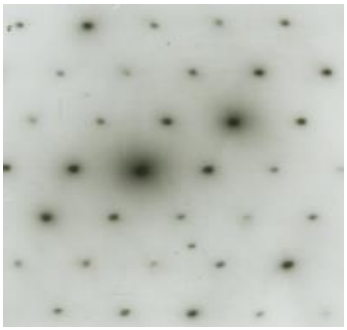

Fig. 2. Typical electronogram of a single crystal that reflects the specific character of its structure, i.e. the presence of the long-range order

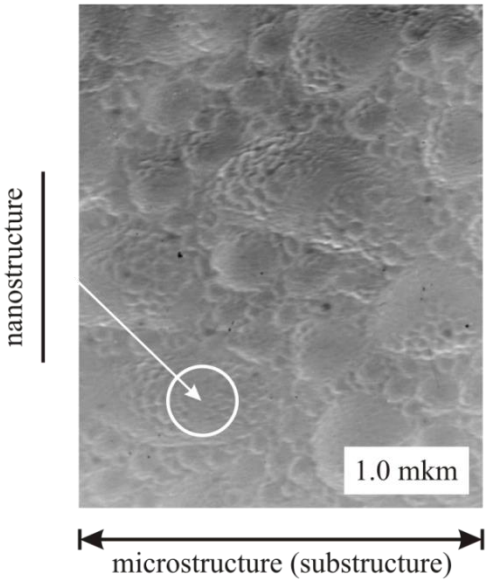

Fig. 3. Transmission electron-microscopic image of the amorphous $As_{40}S_{60}$ film structure at the amorphous matrix level

The second structural level is defined in a form of a continuum of the substance material (fig. 3). We suggest calling it the amorphous matrix structure. Different elements of the amorphous material structure of nano-, micro- and macroscales are revealed on this level.

From our point of view, the structure on the atomic network level must make basis of the amorphous substance definition. Consider the particular examples of such approach presented in other works. Ref. [8] states

that the amorphous solids are not the form of the matter with the long-range order (LRO), they have no large regions with periodical location of atoms and no crystalline microvolumes of the size that allows the Bragg diffraction to be obtained. The author of Ref. [16] notes that the amorphous substance, or the amorphic, is a solid being an alternative to the crystal, it differs from the latter by the lack of translation periodicity and possesses the shear viscosity not less than $10^{14,6}$ P. In accordance with Ref. [17], the term 'amorphous' characterizes the materials with no spatial periodicity of the atomic locations. If there is no LRO in the particle locations along any direction inside the solid, such solid is called the amorphous one. In this case LRO means the availability of periodicity in the locations of the particles. The author of Ref. [18] also considers that the phases characterized by clearly expresses LRO, i.e. the availability of the potential periodic field of the crystalline lattice, belong to the crystalline solids. The phases characterized by only the clearly expressed SRO belong to the liquid and glassy phases. In the gaseous state, the substances have SRO within the limits of the separate molecules only and differ by the instantaneous changes of configurations produced during collisions.

In our opinion, the above points of view do not take into account some important issues. First, combination on the lower hierarchic level of the states located on the higher hierarchic level and belonging to different systems, e.g. to the liquid and solid aggregate states, is inadmissible. Second, there is no sense to define the amorphous substances using the terms not clearly specified. Such formulation simply substitutes the solution of the terminological task by another one. Third, in general case it is difficult, and even most likely impossible, to specify all known and, moreover, unknown yet substances, which could be considered amorphous. Therefore, the available terms must take into account further extension of the class of the amorphous substances in future. Both theoretical and experimental results of the studies allow several criteria common for all amorphous substances to be established right now. Let us analyze critically the properties, which are most frequently suggested as decisive in defining the amorphous materials as the separate class.

1. Isotropic character of the properties. Regarding this criterion, it is necessary first to specify the spatial dimensions of the system, in which it is applied. If these are macrodimensions, then the isotropic character of the properties may frequently reflect not internal essential features of the structure of the substance, by the external manifestation of the peculiarities of its organization into the macrosystem. For instance, on the macrolevel, the polycrystals are isotropic, but they could not be treated as the amorphous substances. On the other hand, a lot of amorphous films demonstrate on the mocrolevel the anisotropy of their properties due to the conditions of directed vapor condensation onto the substrate and due to the properties of its surface structure. If the criterion of anisotropy of the properties is applied to the atomic dimensions, i.e. to the systems with dimension of a few 'unit cells' of the crystalline lattice, it is not proven experimentally that the amorphous substances on that level have isotropic properties. In contrary, high-resolution electron microscopy indicates in many amorphous films and glasses the occurrence of some elements of the translation symmetry and anisotropy of structure on the local level of a few nanometers [19, 20]. In addition, many amorphous films demonstrate such properties (e.g. birefringence), which are anisotropic by their essence [21].

2. Occurrence of the surfaces of arbitrary shape, most frequently, the conchoidal cleavage, at the breakage or cleavage is a criterion that reflects the external properties of the substance and is not the general one, but only that related mainly to glasses. To extend its application over any kinds of the amorphous substances one has to solve a number of problems: what should be considered a 'conchoidal' cleavage of the amorphous film or amorphous ultradisperse particle? Is it possible to consider the cleavage of the single crystal amorphized by the high-energy radiation the 'conchoidal' cleavage?

3. The lack of the local crystalline microvolumes both in the compact and in the disperse state is the criterion of rather the phase homogeneity of the substance than its amorphous essence.

4. The thermodynamical non-equilibrium or metastability of the amorphous substances is often used as their general attribute, and this does not correspond to the minimum of the total energy of the system in these conditions [21, 22]. However, this criterion is not so integrating to cover any possible cases of realization of the amorphous substances. Not all their kinds are kinetically solidified non-equilibrium thermodynamical systems. The amorphous films or the amorphous disperse particles in certain conditions could also be thermodynamically stable systems due to the contribution of the surface energy. In addition, the thermodynamical non-equilibrium itself is a result of manifestation of the specific character of the amorphous substances.

5. Attribution of the amorphous substances to the class of super-cooled liquids [23, 24] means that we have to consider them as the liquid state. Therefore, one has to speak about the amorphous super-cooled liquid. Then, what the crystal is? The crystalline super-cooled liquid? The authors of Refs. [23, 24] also believe that the ordinary glasses could be called the 'glassy liquids', and the molecular crystals with ordered molecule locations in the crystalline lattices and with unordered spatial orientation could be called the 'glassy crystals'. As regards the crystals with orientation order and with unordered location of molecules in the space, they could be called the 'glassy crystallization liquids'. Introduction of the above terms sets a slight wondering. If one continues the analysis in the same direction, the new definitions are obtained. For example, if the 'glassy liquids' exist, then, probably, the 'crystal-like liquids' exist as well; if the 'glassy crystals' exist, then, probably, the 'crystal-like crystals' exist too. In this case the amorphous films could be called the oversaturated condensed vapor. Such novelties give nothing but terminological confusion. Their major shortcoming is a willing to classify the particular substance as belonging to the two aggregate states simultaneously.

6. The lack of the melting point is an important property of the amorphous materials, though the validity of this statement is not established for all amorphous substances of different physical and chemical nature and composition. It should be noted here that a strict generally recog-

nized definition of the melting temperature relates crystals only: the melting temperature is the temperature of the system 'crystalline solid – liquid' in the phase equilibrium state. Thus, this parameter characterizes the equilibrium phase transition to the liquid state of the crystalline substances and its application to the amorphous substances, speaking strictly, is wrong. It is assumed here that with the temperature increase the amorphous substance softens and transits to the liquid state gradually. Indeed, in the case of glasses a continuous transition from the solid to the highly viscous liquid state within a quite wide temperature interval is observed. Such continuous thermodynamical transition is characterized by the melting temperature T_g, not by the vitrification temperature.

In the amorphous films, the melting processes are, as a rule, preceded by rather large-scale structural transformations: crystallization, revaporation or other effects activated by relatively large surface or excess of the defective states. Moreover, in many films the specific pseudophase transition occurs with increasing temperature, i.e. relaxation to the state, which is structurally quite close to glass at the same temperature. In most cases, the above effects do not allow the initial amorphous film to be melted. In the amorphous ultradisperse particles, such processes that mask the melting point are sintering and recrystallization.Thus, the lack of the melting point is quite common property of the amorphous substances. However this could not make basis of their studies, because, first, it does not correspond to its rigorous physical sense, and, second, it characterizes not the solid system, but the peculiarities of its transition to another aggregate state – liquid, demonstrating the specific features and differences of the above transition for the crystalline and amorphous materials. Furthermore, this peculiarity is derived and results from the specific character of the structure of the amorphous substances.

7. The lack of LRO or spatial periodicity in the locations of the structural particles of the substance is a property introduced by the overwhelming majority of researchers as a criterion of defining the amorphous materials. However, this criterion has a serious shortcoming: it is rather of the descriptive, not of the concise character [25, 26].

8. Other specific parameters of the difference of the amorphous substance structure as compared to that of crystals are also often suggested as the definitive ones. For example, they include the stability of the chemical bond lengths and the interbond angles for the crystals as well as the possibility of variation of these values in the amorphous substances [27]. This peculiarity is also quite important, since it indicates the possible causes that specify the lack of LRO in the amorphous substances. However, it is not general enough to make basis for the amorphous substance definition. This relates to other structural parameters as well. In particular, it is emphasized in Ref. [27] that all the chemical bonds in the crystal are saturated, while in the amorphous solid a number of defective broken bonds occur.

9. Certain properties quoted in Refs. [27, 28] also can not be the definitive criteria. Note that in the crystals the energy forbidden band is strictly defined, while in the amorphous solids the localized allowed energy states occur in the middle of the forbidden band; a sharp optical absorption edge is typical for crystals, whereas in the amorphous materials there is an extended tail of the electron states at the absorption edge. In addition, both resistive and blocking contacts are possible for the crystals, while the most of the contacts in the amorphous substances are resistive. Some properties may serve only the distinctive criteria to classify different classes of the amorphous materials, e.g. to differentiate metallic glasses from other types of glasses [29]. All the aforementioned differences are derived from the structure, and their cause must lie in the specific character of different types of the amorphous materials and crystals.

The analysis of the above criteria shows that most of them reflect external, secondary or partial properties of the amorphous materials. Therefore, attempts to define the amorphous substances according to their specific properties are untenable. This is confirmed by the fact that if one takes charge transfer as the indicator of the appearance of the amorphous phase at ion implantation into crystal [30], then it is possible to find the occurrence of the amorphous phase in terms of percolation leaking theory as resulted from the formation of the infinite cluster well before its finding by the structural methods. At the same time, the above definitions in the most cases actually specify the distinctive attributes of the solid state of the substance as compared to the liquid state independent of the fact is this substance amorphous or not.

The lack of periodicity and LRO in the locations of the structural particles is, according to the most of researchers, the specific feature that must form the basis of defining the amorphous solids [8, 14, 25, 31–34]. It should be noted here that the most of the above criteria of pointing out the amorphous materials as a separate class are only the consequence of the LRO presence or absence (table 1).

Note as an example of such approach the following definition of the amorphous substances: the amorphous solid is a solid state of the substance characterized by the isotropic character of the properties and by the lack of the melting point. With increasing temperature the amorphous materials soften and gradually transit to the liquid state. Such behavior is due to the lack of LRO in their structure. It is clearly seen that the definitive properties of the amorphous substances are specified by the lack of LRO.

Thus, only one attribute, i.e. the lack of LRO, is so general and internally inherent in the amorphous solids that today this attribute forms the basis for defining the amorphous substance category by the majority of researchers. However, this criterion has substantial shortcomings. The principal of them is that the character of the diffraction data does not indicate directly the lack of LRO in the amorphous substances. Precise X-ray and neutron imaging experiments for the most of the amorphous substances under study show the radial distribution functions for atoms with maxima extended up to 2–3 nanometers. Such distances correspond to the real nanoobjects with about 5 nm dimensions. In such situations, the question about the lack of LRO is, in our opinion, disputable. Therefore, the above approach should be complemented by the parameters that describe the special character of the real structure of the atomic network in the amorphous materials, i.e. specify the available peculiarities, not only the lack of LRO.

Table 1

Criteria of pointing out the class of the amorphous substances, which directly or indirectly reflect the lack of LRO

Criterion	Specific for amorphous materials:	How the lack of LRO is reflected	Notes
Isotropic structure and properties	Most of the amorphous substances, but not all	Directly	Should be considered on the scale level not exceeding few nanometers
Conchoidal cleavage	Mostly for glasses	Indirectly	Inherent in macrosamples
Lack of the crystalline regions	All amorphous solids	–	Criterion of the single phase, not of the amorphous nature
Thermodynamical non-equilibrium	Most of the amorphous substances, but not all	Indirectly	Result of manifestation of the structure and the external shape of samples
Super-cooled liquid	Mostly for glasses	Directly	Simultaneous combination of two aggregate states is not allowed
Lack of the melting point	Not for all solids	Directly	Characterizes phase transition, not the substance
Lack of LRO	For all substances	Directly	General, but quite descriptive criterion. Requires specification of SRO
Large defective chemical bonds concentration	For most substances	Indirectly	Definition should be on the ideal state level, not real amorphous substances
Peculiarities of specific properties	Could be used to point out separate types of amorphous solids	Directly	Result of the specific features of the structure

In Ref [35], the following definition of glass is suggested: the solid material in which the unit cells (structural units) are bound by the chemical bonds into chains, rings, strips, layers and 3D networks in such a way that the angles between the chemical bonds that link the structural units are not equal, but fixed within the specified boundaries limited by the nature of this substance boundaries leading to the limited variational SRO and to the lack of LRO in the locations of atoms.

This definition corresponds to the modern understanding of the specific character of the amorphous substances. But it has essential drawbacks. First, it provides an extremely broad specification of the character of the SRO construction. Second, this definition restricts the causes of LRO disappearance only to the occurrence of variation of the angles between the bonds that link separate structural units. As a result, this definition loses overall generality necessary to join all the amorphous substances by a single term.

Thus, the diffuse patterns of X-ray, electron and neutron diffraction are the experimental condition of manifestation of the amorphous state in the certain substance. Such patterns are qualitatively similar for a variety of amorphous substances reflecting, thus, the unity of principles of constructing their atomic structure. Therefore, terminological definition of the amorphous solids must relate to the peculiarities of their atomic network structure. The main structural criterion of pointing out the separate class of amorphous materials is the lack of LRO or translation periodicity in the spatial location of the structural particles. However, this criterion must have more rigorous character specifying the causes of LRO disappearance in the amorphous substances. The character of organization of the amorphous SRO must be decisive here.

The analysis of this question requires clear definition of the LRO and SRO notions. No unambiguity and strictness are seen in the approaches suggested by different authors. Consider, for example, some definitions. SRO means the proper location of a certain number of neighbors close to atom, ion or molecule [25]. LRO is the location of the particles in a certain sequence with the formation of a single 3D lattice [17], or, according to Ref' [17], a number of closest neighbors surrounding atom chosen as the central one. Here the main SRO parameters are the number of neighbors (coordination number) and the distances between them (coordination sphere radius). Such SRO is called the coordination order. For liquids that contain atoms of different chemical elements the distance SRO is not a sufficient characteristic. In this case it is necessary to take into account not only the number of atoms that surround the central atom, but also their sort. We shall call SRO with the sorts of atoms being taken into account the sort order. LRO and SRO are the symmetry or correlation in the mutual locations of solid structure elements (atoms, molecules, group of atoms of certain configuration) or the spatial correlation of the physical values (polarizability, magnetic moment) [33].

Table 2.

General methodological principles of styding the amorphous substances

Idea of principle	Atomic network level	Continuum level
Pointing out two qualitatively different structural reasons	Atomic structure in a form of coupled structural particles discretely located in the space	Microstructure and macrostructure of the sample in a form of a continuum
Ideal amorphous structure approximation	Ideal atomic network, all atoms with saturated valences	Ideal amorphous matrix in a form of a permanent homogenous continuum with the uniform distribution of the physical and chemical parameters
Clear resolution of different structural scales	Short-range order. Intermediate-range order. Mesoscopic ordering	Nanostructure. Microstructure. Macrostructure
Inclusion of all structure-defining and structure-specifying factors	The initial state of the substance. Conditions of formation. Action of external factors. Relaxation processes and structural transformations	
Construction of the structural models	The model of atomic network with 2–3 nm dimensions	The models of formation of different-type heterogeneities of the amorphous matrix

When considering the above definitions some comments arise. 1. What does 'proper' location of the nearest neighbors around the atom means? How the 'proper' and 'improper' neighbor location of this atom could be defined? 2. What does 'a certain number' mean? Two, five or hundred nearest neighbors? LRO and SRO can be qualitatively different, e.g., coordination, distance etc. 4. The notions of SRO and LRO relate not only to the atomic

structure of the substance, but also to their different properties. Therefore definition of these notions must be general for all cases, and clear definition of both SRO and LRO must take into account the above comments.

Thus, it seems expedient to hold to several general principles when studying the amorphous substances (see table 2). They are based on the clear distinguishing of the two structural levels. The ideal structure approximation should be used in general definitions on these both levels.

In the case of the real amorphous materials one has also to take into account the strong influence of the conditions of their production on the structural, physical and chemical parameters of the samples produced. Due to the action of these factors, different polymorph modifications and structural states of the amorphous solids even of single-atom substances can be revealed. The latter, in turn, determine different properties of the amorphous samples of the same chemical compositions produced in different technological regimes. Accounting the substantial losses of information, which accompany the most of modern methods of structural analysis of the disordered systems, makes irreplaceable construction of the scale models of the amorphous substances with the number of atoms corresponding to the conditions of application of the laws of statistical mechanics and thermodynamics.

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Боркач Е.И., Иваницкий В.П., Ковтуненко В.С. Терминология и определение аморфных веществ

Аннотация. Экспериментальным условием установления аморфности состояния любого вещества есть диффузные картины дифракции рентгеновских лучей, электронов и нейтронов. Такие картины качественно одинаковы для самых разнообразных по своей природе аморфных веществ, что отражает единство физических принципов построения их атомной структуры. Поэтому и терминологическое определение аморфных тел должно основываться на особенностях структуры их атомной сетки. Главным структурным критерием выделения класса аморфных веществ есть отсутствие у них трансляционной периодичности пространственного расположения структурных частиц или дальнего порядка. Но этому критерию необходимо придать более строгую основу путем конкретизации причин исчезновения в аморфных материалах дальнего порядка. И эти причины определяются характером организации аморфного близкого порядка.

***Ключевые слова**: аморфные вещества, неупорядоченные системы.*