

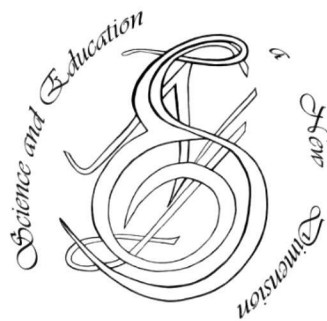
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CONTENT

MEDICINE AND BIOLOGY	7
<i>Komisar O.S., Boyko M.F.</i> Heavy metals in gametophytes of moss <i>Bryum caespiticium</i> Hedw. in parks and on Lenina Avenue in Mykolayiv city (Ukraine)	7
<i>Boiko M.F., Khodosovtsev O.Ye., Gavrylenko L.M., Melnyk R.P., Klymenko V.M., Shaposhnikova A.O.</i> Phytodiversity and Lichenodiversity in the Conditions of Agricultural Landscapes In Southern Ukraine	11
<i>Дуло О.А., Фурман Ю.М.</i> Вивчення рівня фізичного здоров'я дівчат гірських районів Закарпаття за метаболічним рівнем анаеробного енергозабезпечення	15
<i>Лизогуб В.С., *Макаренко М.В., Юхименко Л.І., Хоменко С.М., Коваль Ю.В., Кожемяко Т.В.</i> Вікова динаміка сенсомоторних функцій людей із слуховою депривацією	20
<i>Макарчук М.Ю., Глазирін І.Д., Смоляр С.І.</i> Особливості біологічного дозрівання учнівської молоді жіночої статі визначеного за темпами статевого розвитку	25
<i>Рудишин С.Д.</i> Еволюція екосистем в контексті сучасної природничо-наукової картини світу	30
<i>Тимофійчук І.Р., Анохіна С.І., Кузнєцова О.В., Боштан С.В., Семененко С.Б., Слободян К.В.</i> Зміни гормональної активності та біохімічні перебудови кори наднирникових залоз на тлі розвитку експериментального цукрового діабету у щурів різних вікових груп	35
<i>Черненко-Курагіна Н.П.</i> Індивідуальні реакції гемодинаміки головного мозку та регуляції серцевого ритму при розумовій діяльності з низькою швидкістю пред'явлення інформації	38
ECOLOGY	43
<i>Malyarenko O., Samoilenko V.</i> Regional ecological networks: developed geoinformation modeling approaches	43
VETERINARY SCIENCE	47
<i>Maksymovych I., Slivinska L., Winiarczyk S., Buczek K., Staniec M.</i> Hematological and serum biochemical reference values in healthy working horses Hutsul breed	47
<i>Hudyma T.M., Slivinska L.G.</i> Therapeutic efficacy of Hepatiale®Forte in treatment of the dogs with hepatodystrophy	51
<i>Lukashchuk B.O., Slivinska L.G.</i> Prophylactic effectiveness of phytobiotic feed additive for non-contagious diseases of the gastrointestinal tract in suckling piglets	54
CHEMISTRY	57
<i>Брем В.В., Кожухар В.Я., Іванченко Л.В., Дмитренко І.В., Буга С.П.</i> Відбір проб рідкого флюсу для газового аналізу на вміст водню	57
ASTROPHYSICS	63
<i>М.Yu. Skulsky</i> On the wave structure in the spatial organization of the Solar planetary system	63

PHYSICAL AND MATHEMATICAL SCIENCES	68
<i>Borkach E.I., Ivanitsky V.P., Kovtunenکو V.S., Ryaboschuk M.M.</i> Glasses and amorphous films as two kinds of non-crystalline state of substance	68
<i>Петрецький С.І, Міца В.І, Голомб Р.І, Кондрат О.І, Попович Н.І, Боркач Є., Мартон А.І, Вереш М., Чік О.</i> Синхротронні фотоелектронні спектри, склад та локальна координація атомів в плівках Ge_2S_3 при введенні миш'яку	73
<i>Сагітов Ю.Х.</i> Гибридное интегральное преобразование свёртки: основная теорема, следствия из неё, некоторые приложения	77
ARCHITECTURE AND CONSTRUCTION	81
<i>Sadowska E.J.</i> The late baroque wooden church of Michael the Archangel in Szalowa(Poland) – the church from wooden model	81
<i>Шульга Г.М.</i> Методика моделювання планировочної організації територіальних систем рекреації в Українських Карпатах	87
TECHNICAL SCIENCES	91
<i>Дячок В.В., Гуглич С.І.</i> Математичне моделювання біологічних процесів ускладнених масопереносом	91
<i>Зав'ялов В.Л., Мисюра Т.Г., Бодров В.С., Запорожець Ю.В., Попова Н.В., Деканський В.Є.</i> Закономірності дії пульсуючих потоків в умовах протитечійного розділення фаз при віброекстрагуванні із рослинної сировини	95
<i>Ковальський Б.М., Занько Н.В., Шовгенюк М.В.</i> Визначення колірних характеристик друкарських фарб за спектральними кривими відбивання	100
<i>Небылица Ю.Н.</i> Интеграция системы экспресс-диагностики процесса электроэрозионного проволочного вырезания в технологическое оснащение	104

PHYSICAL AND MATHEMATICAL SCIENCES

Borkach E.I., Ivanitsky V.P., Kovtunenکو V.S. Ryaboschuk M.M. Glasses and amorphous films as two kinds of non-crystalline state of substance

Borkach E.I., CSc in Physics, associated professor

II. Rákóczi Ferenc Transcarpathian Hungarian Institute, Beregovo, Ukraine

Ivanitsky V.P., Dr. Science in Physics, Prof., Uzhgorod National Universit, Uzhgorod, Ukraine

Kovtunenکو V.S. CSc in Physics, associated professor, Cherkasy State Technological University, Cherkasy, Ukraine

Ryaboschuk M.M., associated professor, Uzhgorod National Universit, Uzhgorod, Ukraine

Abstract. It is shown that the glasses are a kind of the amorphous state produced due to the reverse phase transition 'liquid–solid' under the external conditions providing the suppression of the crystallization processes. The structure of the glass is determined by the structure of the initial melt and the character of the processes of its evolution during vitrification. The amorphous films are the type of the amorphous state produced as a result of irreversible processes of condensation of the vapor and plasma flows onto the substrates under the surface conditions that exclude crystallization processes. The atomic and molecular composition, the energy condition of the particles of the vapor and plasma flows and the character of their condensation onto the substrate surface appear to be decisive for the amorphous film structure. Production of glasses and amorphous films from different aggregate states of the substance stipulates substantial differences of their structure and all specific features and properties.

Keywords: *amorphous materials, glasses, amorphous films*

Presence of short range order and absence of long range order combine different types of non-crystalline state with substantial distinctions into one group. Among the class of the amorphous solids, a group of the substances produced by melt cooling is very wide and especially important in practice. The process of their formation starts from the equilibrium liquid state that exists at the certain temperature and pressure. Further temperature and time regimes of fast cooling with solidification modify, to some extent, the structure of the initial melt. Nevertheless, in the solid amorphous state these substances reproduce necessarily the specific peculiarities of the structure of corresponding initial melts. As a result, this group of substances has a good reason to be clearly distinguished of a wide class of the amorphous solids into the separate subclass of glasses [1].

Considering the glasses and the glassy state as one of the types of non-crystalline solids is traditional in modern physics of the condensed substances [2-5]. One should note here that the term 'glass' is lower in hierarchy than the term 'non-crystalline', while the terms 'vitrification' (for inorganic materials) or 'gumming' (in organic materials) are treated as the basic kind of the amorphous state formation processes. At the same time it is assumed that the terms 'non-crystalline' or 'amorphous' are more general as compared to the term 'glassy', and the class of the amorphous solids is much wider and comprehensive than the class of the glassy bodies – like the general is wider than the particular. It is also noted that the glasses are always amorphous, but not all of the amorphous substances are the glasses. Expansion of the term 'glass' to the level of the 'non-crystalline solid' or 'solid amorphous substance' is not feasible. The statement that the crystal can be transferred to the glassy state by irradiating it by high-energy particles without melting could not be considered correct as well [6].

The glassy substances have some similarity with the supercooled liquids [7]. This is due to the fact that vitrification is the continuous second-order phase transition from the supercooled liquid to the solid state at the vitrification temperature. The glass and the supercooled liquid correspond to the two close but different thermodynamical states separated by the phase transition. Both these

states are disordered, but the nature of disordering in them is different. The homogeneous liquid is the state with the equilibrium structural fluctuations, whereas in glasses these fluctuations are frozen. The above two states are similar by their atomic structure [4]. In most cases they are similar by the isotropic character of their properties. The peculiarities of the structure of the crystalline phases in this substance influence considerably formation of short range order in liquids.

When defining the term 'glass', the principal difficulties arise due to the gradual character of the change of all characteristics of the glassy substance at the transition from the liquid state to the glassy one and vice versa. The most complicated is the problem of determining the boundary between the liquid and the glass. To solve this problem one has to begin with the most general definition of the terms 'glass' and 'glassy state' and then turn to the method of finding the quantitative distinctions between the liquid and the glass. According to Ref. [8], the 'glassy substance is the amorphous solid produced by cooling the liquid (viscous melt) without crystallization'. Note that equality of the notions 'amorphous melt' and 'metal glass' indicated in Ref. [9] is due to the singleness of the notions 'melt' and 'glass'. This singleness results from the same initial liquid state, which the above substances are produced of. The following properties are considered characteristic for the glasses:

1. Their formation results from cooling (freezing) the liquid without crystallization or at the pressure increase. That is, the liquid state is the initial state for producing glasses, while the process of glass formation is the vitrification phenomenon [10, 11–14].

2. The process of the transition from the liquid state to the glassy one must be reversible, i.e. the process reverse to vitrification must be realized, i.e. softening [1-3, 15] or defrosting [16, 17].

3. The lack of the characteristic vitrification temperature T_g and the corresponding transition at this temperature are the essential features that distinguish the amorphous state from the glass. In many amorphous materials, such transition can be masked by the crystallization processes initiated by the excess of the defective states or by

the surface. Other amorphous materials, in particular, the films, can demonstrate the pseudo-transition with certain temperature close to T_g : the structural relaxation to the state identical to the liquid cooled to this temperature. After this transition or simultaneously with this transition the above material crystallizes.

4. In the isobaric-isothermal conditions in the glass, the time of relaxation of any degree of freedom to equilibrium is too large as compared with the duration of the experiment or of the glass use [10, 18]. The glass samples must have macroscopic size at least in one dimension [15], and this attribute, probably, distinguishes the glass from ultradisperse particles.

The glassy state is thermodynamically stable [19] and possesses the mechanical properties of solids [20].

The glass has the properties of undefined chemical compounds and possesses the isotropic properties in the macroscale, while fragility and conchoidal cleavage are also typical for it [2].

Which of the above properties became generally accepted and most frequently used definitions of the glass? One of them is the following definition [21]: 'The glasses are the amorphous solids produced by the melt supercooling independent of the chemical composition and temperature region of solidification, which due to the gradual increase of viscosity possess mechanical properties of the solids; the process of the transition from the liquid state to the glassy state must necessarily be reversible'. 'The glassy state is the type of the amorphous state, in which the substance has the dynamical viscosity coefficient above 10^{12} – 10^{13} P, i.e. it has mechanical properties similar to those of the crystalline solid and differs by the ability to return after being melted to the initial state in the predetermined cooling mode. The glass is a monolithic material consisting primarily of the glassy substance'. In these, quite similar, definitions, the following properties are chosen from all the above ones: a) the glass is one of the kinds of the amorphous substances; b) it is solid; c) the liquid is the initial aggregate state for glass production; d) the reversibility of the transition to the liquid state, i.e. the reversibility of vitrification and softening processes, is inherent in glasses.

In this relation, it is difficult to agree with the point of view of G.M. Bartenev and D.S. Sanditov [19], according to which it is almost impossible to give definition of the glass, which will cover all possible compounds and conditions of their formation. Definition of the glass, which is valid at least today, is given above. If it appears unacceptable for a certain state of the substance, this means that the above substance belongs to the other amorphous state. In case one finds new, more general, attributes or properties of the glassy state of the substance, the definition of the category of glass will be changed as well.

Note that, according to Ref. [6], the solid body differs from the liquid, first, by the lack of fluidity and, second, by the lack of the changes in the relative locations of atoms. This definition allows the solid state of the substance to be clearly distinguished from the liquid state at the change of the aggregate state, i.e. at the crystallization or melting. In the process of vitrification, at the continuous variation of the fluidity and relative mobility of the structural particles, it is quite difficult to find the boundary between the liquid and solid states. This could be made only on the basis of some general agreement. As a result, the author of Ref. [6]

reports the following definitions. Vitrification is the transition from the liquid state to the glassy state due to the freezing of the structural (configuration, conformation) fragments of the liquid that accompanies the temperature decrease or the pressure increase. The glassy state is the amorphous non-equilibrium state, to which the liquid transits as a result of cooling or increasing the pressure acting on the liquid. It differs by that one cannot neglect the effects related to approaching the equilibrium by the substance during the period of observation or operation.

However, speculations [6] are not completely correct for two reasons. First reason is a simplified consideration of the vitrification process as a simple structural fragments freezing in the liquid under the temperature decrease. In reality, the process of vitrification is more complicated and includes both freezing of the structural changes in the liquid and the simultaneous processes of the solid phase nucleation [22]. Second, high-temperature phase tempering indeed suppresses weak diffusion processes, however, it does not interfere the reconstruction in a wider scale. Furthermore, quick cooling is frequently used not to fix the high-temperature variety of the structures but, in contrary, to initiate new global transformations. Examples are the steel tempering into the martensite, the alloy tempering on the copper and manganese bases to provide them with the shape memory properties. The statements that the glass is a solidified liquid and that the liquid is a softened glass contain the tautology inefficient for the researcher and technologist [22]. There the glass should be considered not the frozen liquid but the result of the synergetic phenomena of the system self-organization in the course of the change of the structure in the strongly cooled medium, i.e. due to the evolution of the melt structure from 'existing to emerging' [23]. As a result, vitrification should be defined as more general notion located at the same level with the notion of crystallization. Since crystallization is the phase transition from the liquid or gaseous phase to the crystalline one, then vitrification is the transition from the liquid phase to the non-crystalline one. The decisive role in this transition belongs to the kinetic phenomena [24]. Here the structural peculiarities of the melt that force the latter to transform into the glass, not the crystal, is the unsolved problem, i.e. the glasses are produced of the liquids but are formed at the melt structure freezing in the processes, which are tendentially directed toward the crystal formation.

Both the crystalline and non-crystalline solid states are formed not only from the liquid but also from the gaseous phases. Most frequently the films or coatings with the thickness from monoatom layers to dozens of micrometers are formed at the gas-phase deposition.

The amorphous film state of the substance is terminologically unclearly determined in literature like the notion of the amorphous state. For example, according to one of the points of view, the thin films are considered the disperse form of the amorphous substance [25].

The amorphous films should be considered a kind of the amorphous solid state [1] produced by the direct condensation of the gas or plasma flows. The phase composition and the structure of the films substantially depend on the intensity of the atomic, molecular or ion plasma flows. It should be noted here that, as follows from the above definition, the difference between the glasses and the amorphous films could not be reduced to different bulk and surface states of

the substance only. The glasses could also be realized in a form of the thin (glassy) films), for instance, at the super-fast tempering of the metal melts or at the condensation according to the 'vapor-liquid-glassy film' mechanism [26]. Such glassy films must be similar by their properties to the glasses more or less modified by the influence of the surface and the peculiarities of their production.

The degree of microheterogeneity could not be the attribute of the glassy substances. Therefore the comments [9] stating that the glass is the homogeneous state of the substance and the amorphous sample is microheterogeneous state are incorrect. As an example of such incorrectness one may note the comparison of the single crystals, polycrystals, block crystals, ultradisperse particles, solid solutions, inhomogeneous glasses after liquation and so on.

It is generally assumed that if the same substance is produced in a form of the massive glass and the amorphous film, then its properties are quite close [27]. For example, the short range order characteristics in them almost coincide [28]. The only exception is the properties, which are closely related to the specific character of production of these two kinds of the amorphous substances.

Let us analyze the distinctive features of the condensed amorphous film state.

1. The methods of production of the amorphous films are related to the use of the phase transitions between the aggregate states 'gas (plasma) – solid' with condensation of the latter onto the relevant substrates.

2. A specific, close to two-dimensional, geometry of amorphous films, in which the directions parallel and perpendicular to the substrate are essentially non-equivalent.

3. Strong manifestation of the effects of the surface inter-phase interactions at the boundaries 'substrate-film' and 'film-environment' [29, 30]. In many cases the amorphous films must be considered a complex inseparable system 'substrate-film-environment'.

4. The higher degree of the thermodynamical non-equilibrium (metastability) of the amorphous films as compared to the glasses. This is revealed in the considerable differences of the transition processes in the amorphous films and glasses to more equilibrium structural states and at their crystallization [31]. The relaxation time at the glass synthesis is larger than that at the amorphous film condensation [32] and, therefore, the structure of the glass is more equilibrium.

5. A large intensity of relaxation changes of the structure and the properties in the post-condensation period.

6. The influence of different dimensional effects (including the phase and quantum effects) on the processes of structure formation and on the physical parameters [33–35].

7. Due to a larger non-equilibrium and multiple-factor character of condensation processes, the concentration regions of production of the stable amorphous films in the complex systems are, as a rule, much wider than the corresponding vitrification regions.

8. Various atomic configurations of the amorphous structural network are realized in the wider ranges [36].

9. The possibility of the multiple reverse transitions to the melted state [27], which are the specific attribute of the glass, is completely excluded in the amorphous films. Such transitions of the films to the liquid state are masked or hindered by the processes of the structural relaxation

and crystallization in the films. This peculiarity is not general and could be violated in some cases. Furthermore, a considerable difference in the crystallization of the amorphous and glassy materials reflects the fact that we deal with different states of substance. Respectively, they will demonstrate the temperature and the time dependences of the physical and chemical properties and the character of the phase transitions.

At the same time, one has also not to overemphasize the above distinctions, as is suggested, for example, in Ref. [37]. It is stated that one may compare only the glasses and single crystals, the amorphous and the polycrystalline films of the same chemical composition. Note that the chemical individuality of each particular substance is revealed differently in its different realizations. However, comparing the specific character of the glasses and amorphous films, one may note that the films in many points are similar to the glassy state, though they possess a series of specific features [38]. The main distinctions are stipulated by the character of the initial state of their formation, i.e. by the flow of the vapor or plasma particles.

It is evident that the atomic, molecular and ionic composition of the above flows will substantially stipulate the structure of the films produced. At the same time, an important role in the structure formation is also played by the energy state of the particles of the vapor and plasma flow and by the kinetics of the processes of their condensation on the substrate surface. In this case, contrary to vitrification, the character of formation of the amorphous films structure during their condensation is studied extremely insufficiently. In general, the action of all these factors must ensure the considerable difference of the structure of the amorphous films and glasses of the same substance.

In the structural relation, it is assumed [39] that the films include large concentration of the homobonds, whereas in glasses they are almost absent. One has to take into account here that the homobonds themselves may enter different structural formations in the films and glasses (e.g., separate clusters or continuous network). In this relation, the films are considered more defective by the degree of chemical binding and by the type of the chemical bonds [40]. Experimentally such distinctions are revealed in the electronic properties of the films and glasses. Consider, as example, Ref. [41], where it is shown that the specific electric conductance of the amorphous GeS₂ films is four orders larger than that in the relevant glasses of the same composition. In addition, even film annealing at the temperatures close to T_g does not lead the value of their conductance to the characteristic values for glass. This means that the glasses and the films differ not only by the concentration of the defects but also by the peculiarities of the atomic network structure, i.e. by different geometric topology of the structural units (SU) and their relation.

One more important moment is that the surface regions of the film will strongly differ from the bulk ones. In particular, large densities of free chemical bonds may occur at the surface and their considerable fraction may take part in the surface structure reconstruction. Some of the film properties will depend on the film thickness. A great role could be played in films by the surfaces related to the internal pores and cavities. Moreover, other essential differences in the amorphous film and glass structure of the same substance are possible.

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Боркач Е.И., Иваницкий В.П., Ковтуненко В.С. Рябошук М.М.

Стёкла и аморфные пленки как два вида некристаллического состояния вещества

Аннотация. Стекла являются собой один из видов аморфного состояния, который образуется в результате обратимого фазового перехода жидкость – твердое тело при таких внешних условиях, которые обеспечивают подавление процессов кристаллизации. Структура стекол определяется структурой исходного расплава и характером процессов его эволюции во время стеклования. Аморфные пленки – это вид аморфного состояния, который формируется в результате конденсации паро-плазменных потоков на подложках в таких условиях, которые исключают инициирование процессов кристаллизации. Определяющими факторами для структуры аморфных пленок есть атомно-молекулярный состав и энергетическое состояние частиц паро-плазменных потоков, а также характер их конденсации на поверхности подложки. Получение стекол и аморфных пленок из разных агрегатных состояний веществ обуславливает существенные различия их структуры и всех физико-химических свойств.

К основным отличительным чертам аморфного пленочного состояния можно отнести: а) получение в результате протекания фазовых переходов газ (плазма) – твердое тело с конденсацией на соответствующих подложках; б) особая, близкая к двумерной геометрии, в которой направления параллельные и перпендикулярные подложке существенно неэквивалентные; в) сильное проявление эффектов поверхностного межфазного взаимодействия на границах подложка – пленка и пленка – внешняя среда; г) более высокая степень термодинамической неравновесности (метастабильности) в сравнении со стеклами; д) интенсивное протекание релаксационных изменений структуры и свойств в послеконденсационный период; е) существенное влияние разных размерных эффектов на процессы структурообразования и физические параметры; ж) более широкие концентрационные области получения для двух- и более компонентных систем; з) более широкий диапазон реализующихся атомных конфигураций ближнего и промежуточного порядка неупорядоченной атомной сетки; и) невозможность многократных обратимых реверсивных переходов в исходное (парообразное) или жидкое состояние.

Ключевые слова: аморфные вещества, стекла, аморфные пленки