УДК 621.315.592

M. I. Mar'yan, A. A. Kikineshy, A. Szasz¹ Self-organizing processes and dissipative structure formation in the non-crystalline materials

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The dissipative structure formation in the non-crystalline materials is related to the selforganizing phenomenon. Within of a given approach non-crystalline states is considered as an outcome of the previous self-organization, during which may promote the minimum energy dissipation and facilitate the technological process.

Keywords: synergetics, self-organization, non-crystalline state.

Стаття поступила до редакції 11.06.2001; прийнята до друку 12.06.2001

I. Introdoction

ideas of the non-equilibrium The thermodynamics predicting the possibility of the non-crystalline structure with new unique properties non-equilibrium in strongly conditions have been widely developed. In this case by the structure the method of the element organization and the spatial-temporal character of their intercorrelation are meant. The dissipative structure formation in the noncrystalline materials is related to the selforganizing phenomenon.

The synergetic approach to the shaping and exposition of non-crystalline structure enables to aggregate the thermodynamic and kinetic concepts so that they do not exclude, but on the contrary, self-consistent supplement each other [1-5]. Within the framework of a given approach non-crystalline states are considered as an outcome of the previous self-organization, during which may promote the minimum energy dissipation.

By the term "non-crystalline substance" the structural disruptions are meant most frequently being manifested as the absence of the correlation of the physical quantity, which describes the present system at the distances defining the disordering scale. The noncrystalline solid state is usually considered as a metastable one, which does not correspond to the minimum of the total energy and will transit in time to the stable equilibrium state. From this standpoint the total order is the one that appears due to the non-vanishing correlation of all physical quantities across the total volume of the system. That approach to the non-crystalline solids is in the same relation and has the same specific features as, for example, the flat picture and the real three-dimensional image. Historically, such an approach to noncrystalline materials has been realized due to the description of the initially crystalline spatially ordered solids and the attempt to extend the results for the crystals to the disordered substances. The aforementioned consideration lays the "secondary" relation to the non-crystalline materials, when the essential peculiarities of their operation are rejected. In our opinion, the situation would change

drastically if the structure and properties of the condensed media were started initially just from non-crystalline materials and the crystalline media were treated as a special case of more common non-crystalline ones. In this case a host of phenomena observed in noncrystalline materials would allow one to catch the general hypotheses and models which belong to the unified approach based on the ideas of synergetics [1,2]. One can formulate the following concepts of non-crystalline solid state [2,5]:

- any substance can exist in the noncrystalline solid state in one or another form;
- non-crystalline materials are the dissipative structures formed as the method of system organization;
- non-crystalline substances are characterized by the criteria which define the domain of their stability in this state, i.e. the boundaries of the stability with respect to the control factors, which affect the system (i.e. the temperature, pressure, radiation and the rate of their variation, etc.);
- the role of non-linear modes in the formation of the non-crystalline non-equilibrium stationary system of any degree of complexity is equal and versatile;
- the main parameters, which specify the noncrystalline system, are the spatial-temporal correlation of physical quantities describing it and the life-time of the dissipative structure;
- the general regularities established for the non-crystalline state are also valid for the crystalline one.

II. Model of system

As the temperature of the solid is elevated to the melting temperature, the concentration of point defects (of the internodal atom (vacancy) type) or extended defects (of the dislocation type) increases sufficiently. Such defects taken separately are not the disordered phase nuclei. However, if they are accumulated within a certain macroscopic domain, then such area of disordering corresponds to the structure inherent to the soft states, i.e. it has a shearing level dynamic instability. а high of displacements and the instability related to the defect formation. Hence, within the phase transformation interval along with homogeneous fluctuations (i.e. the thermal displacements of atoms) the effect of heterophase fluctuations increases, being the formation of the liquid-like macroscopic domains inside the solid-phase state under study [6].

The "soft" states are specific structural states with an additional degree of freedom in the configurational space. These states exist due to the spatial fluctuations of the short-range order parameters (the number and the length of the bonds, the interbond angles) near their average values and the formation of the defect states in the highly non-equilibrium conditions. They are intrinsic for non-crystalline solids. Atoms in the "soft" states possess a considerable level of static displacements and, consequently, the vibrational anharmonism and the ability to the spatial rearrangements. The bonding topology and the configurational parameters which describe the "soft" states, are accounted for and detailed in terms of certain structural models. We shall analyze the general tendencies of the formation of the macroscopically ordered structures in the noncrystalline solids away from the equilibrium state. Therefore, we shall use the universal characteristics of the "soft" states.

Now a reasonably large number of the experimental data are available allowing one to state that the structural formations of a certain chemical composition may exist within the $T < T_m$ temperature range as well as in the melt or in the non-crystalline solid [7]. The presence of heterophase fluctuations is confirmed by the studies of the semiconductor melts, the viscosity coefficient and the internal friction at the vitrification of liquid as well as by the Mossbauer spectroscopy, Raman spectroscopy and the X-ray structural analysis data [8-10]. The probability of the heterophase fluctuation formation and the relationship between various microscopic states are defined by the conditions of equilibrium the interphase for the equilibrium phase transformation [11-14] and by the degree of deviation from the equilibrium for highly non-equilibrium state transformations [5].

$$H = \sum_{l=1}^{N} \sum_{f} T_{f}(l) \sigma_{f}(l) + \frac{1}{2} \sum_{l \neq l'} \sum_{f, f'} \Phi_{ff'}(\vec{r}_{ll'}) \sigma_{f}(l) \sigma_{f'}(l'),$$
(1)

where $T_f(l) = \frac{\vec{P}^2(l)}{2M}$ is the kinetic energy of the *l*-th atom, $\vec{P}(l)$ is the momentum of the *l*-th atom, $\Phi_{ff'}(\vec{r}_{ll'})$ is an interatomic interaction potential, \vec{r}_{ll} is the interatomic distance and $\sigma_f(l)$ characterises two sets f of locally observed microscopic states: $\sigma_{f}(l) = \begin{cases} 1, \text{ if the atom } l \text{ is in the state } f; \\ 0, \text{ if the atom } l \text{ is not in the state } f. \end{cases}$

The possibility of the formation of the liquid-like states in the solid is, consequently,. $\sigma_{\rm f} = \langle \sigma_{\rm f} \rangle = \frac{N_{\rm f}}{N}$. Let us introduce the effective Hamiltonian $\tilde{\rm H}$ of the system:

$$\widetilde{H} = \sum_{f} H_{f} + (1 - \sigma_{2})^{2} \sum_{l,l',\alpha} \widetilde{I}_{\alpha}(l,l') v_{\alpha}(l), \quad v_{\alpha}(l) = \sum_{m} v_{\alpha} \left(\frac{l}{m} \right) \sigma_{2}(m), \quad (2)$$

$$H_{f} = \sigma_{f} \sum_{l} T_{f}(l) + \frac{\sigma_{f}^{2}}{2} \sum_{l,l'} \sum_{\alpha,\beta} \left[\Phi(\vec{R}_{ll'}) + \frac{1}{2} u^{\alpha}(l,l') \widetilde{\Phi}_{f}^{\alpha\beta}(l,l') u^{\beta}(l,l') \right],$$

where $\widetilde{\Phi}_{f}^{\alpha\beta}(l,l'), \widetilde{I}_{\alpha}(l,l')$ is the self-consistent potential interaction, $\vec{r}_1 = \vec{R}_1 + \vec{u}_1 + \vec{v}_1$ is a radius-vector of *l*-th atom, R_l is the average equilibrium position of the *l*-th atom, \vec{u}_l and \vec{v}_i are the dynamic and the static displacements of the *l* -th atom, respectively, $u_{\alpha}(l, l') = u_{\alpha}(l) - u_{\alpha}(l')$ are the relative displacements, $v_{\alpha}(l/m)$ are the static displacements of the *l*-th atom when the *m*-th atom is in the soft state. The model parameters, i.e. the fraction of atoms in the liquid-like states σ_f , the force constants $\widetilde{\Phi}_{f}^{\alpha\beta}(l,l')$ and the mean-square relative displacements of atoms $y_{\alpha\beta}(l,l') = \langle u^{\alpha}(l,l') \cdot u^{\beta}(l,l') \rangle$ in equation (2) are calculated on the basis of the variational principle for the free energy functional:

$$F \le F_0 + \left\langle H - \widetilde{H}_0 \right\rangle_0, \ F_0 = \sum_f F_{0f} - TS, \ F_{0f} = -\Theta \ln\left\{SP(e^{\frac{-H_f}{\Theta}})\right\}.$$
(3)

Here F_0 is the free energy functional for the effective Hamiltonian,

$$S = -k_{B} \ln \left[\prod_{f} \left\{ \frac{g_{f}!}{N_{f}! (g_{f} - N_{f})!} \right\} \right]$$

state f, $\Theta = k_B T$. The variation of *F* over the parameters $\sigma(\sigma \equiv \sigma_2)$, $\widetilde{\Phi}_{f}^{\alpha\beta}(l,l')$, $y_{\alpha\beta}(l,l')$ at fixed temperature *T* and pressure *P*:

is the entropy, g_f is a statistical weight of the

$$\delta \mathbf{F} = \left[\frac{\partial \mathbf{F}}{\partial \sigma}\right]_{\mathbf{y}_{\alpha\beta}, \tilde{\boldsymbol{\Theta}}_{\mathbf{f}}^{\alpha\beta}} \cdot \delta \sigma + \left[\frac{\partial \mathbf{F}}{\partial \tilde{\boldsymbol{\Phi}}_{\mathbf{f}}^{\alpha\beta}}\right]_{\sigma, \mathbf{y}_{\alpha\beta}} \cdot \delta \tilde{\boldsymbol{\Phi}}_{\mathbf{f}}^{\alpha\beta} + \left[\frac{\partial \mathbf{F}}{\partial \mathbf{y}_{\alpha\beta}}\right]_{\sigma, \tilde{\boldsymbol{\Phi}}_{\mathbf{f}}^{\alpha\beta}} \cdot \delta \mathbf{y}_{\alpha\beta}$$
(4)

allows one to determine their temperature

dependence for the equilibrium $(\delta F = 0)$ and

non-equilibrium ($\delta F \neq 0$) transformations.

Let us find the functional equations to determine the probability of the liquid-like

states formation, force constants and meansquare displacement of atoms. Using equations (3) and (4) for σ :

$$\begin{split} \delta F = & \left[\frac{\partial F}{\partial \sigma} \right]_{y_{\alpha\beta}, \widetilde{\Phi}_{f}^{\alpha\beta}} \cdot \delta \sigma + \left[\frac{\partial F}{\partial \widetilde{\Phi}_{f}^{\alpha\beta}} \right]_{\sigma, y_{\alpha\beta}} \cdot \delta \widetilde{\Phi}_{f}^{\alpha\beta} + \left[\frac{\partial F}{\partial y_{\alpha\beta}} \right]_{\sigma, \widetilde{\Phi}_{f}^{\alpha\beta}} \cdot \delta y_{\alpha\beta} \,, \\ F &\leq F_{0} + \left\langle H - \widetilde{H}_{0} \right\rangle_{0}, \ F_{0} = \sum_{f} F_{0f} - TS, \ F_{0f} = -\Theta \ln \left\{ SP(e^{\frac{-H_{f}}{\Theta}}) \right\}, \end{split}$$

we find:

$$\frac{\partial F}{\partial \sigma} = \frac{\partial F}{\partial \sigma} + \frac{\partial}{\partial \sigma} \Big\rangle H - \widetilde{H} \Big\langle_{_{0}} = \widetilde{T}_{_{2}} - \widetilde{T}_{_{1}} + 2\sigma \Big(\widetilde{\Phi}_{_{2}} - \widetilde{\Phi}_{_{1}} \Big) - 2\widetilde{\Phi}_{_{1}} - 4\widetilde{I} (1 - \sigma) \sigma - 2IN \ln \frac{\frac{g_{_{2}}}{\sigma}}{\frac{g_{_{1}}}{1 - \sigma}} \Big|$$
(5)

Here the following notation has been introduced: *F* is the free energy functional for the effective Hamiltonian, *S* is the entropy, g_f is a statistical weight of the state f, σ_f is the fraction of atoms in the liquid-like states,

 $\widetilde{\Phi}_{f}^{\alpha\beta}(l,l')$ is the force constants and the meansquare relative displacements of atoms $y_{\alpha\beta}(l,l') = \langle u^{\alpha}(l,l') u^{\beta}(l,l') \rangle$, $\Theta = k_{B}T$,

$$\widetilde{T}_{\rm f} = \sum_{\rm l} \left| \frac{\vec{P}_{\rm f}^{\,2}(l)}{2M} \right|, \quad \widetilde{\Phi}_{\rm f} = \frac{1}{2} \sum_{\rm l,l'} \left| \phi_{\rm f}(l,l') \right|, \quad \widetilde{I} = \left| \sum_{\rm l,l'} \sum_{\alpha,m} I_{\alpha}(\vec{r}_{\rm ll'}) v_{\alpha} \left(\frac{l}{m} \right) \right|.$$

Similarly, the functional equations with respect to the quantities $y_{\alpha\beta}(l,l')$ and

$$\widetilde{\Phi}_{f}^{\alpha\beta}(l,l')$$
 are found:

$$\frac{\partial F}{\partial y_{\alpha\beta}(\mathbf{l},\mathbf{l}')} = \frac{\partial}{\partial y_{\alpha\beta}(\mathbf{l},\mathbf{l}')} \Big\rangle H - \widetilde{H} \Big\langle_{_{0}} = \frac{1}{4} \Big(\Big\rangle \nabla^{_{H}}_{_{H'}} \nabla^{_{\beta}}_{_{H'}} \Phi_{_{f}}(\vec{\mathbf{r}}_{_{H'}}) \Big\langle \sigma^{^{2}}_{_{f}} - \widetilde{\Phi}^{_{\alpha\beta}}_{_{f}}(\mathbf{l},\mathbf{l}') \Big\rangle, \tag{6}$$

$$\frac{\partial F}{\partial \widetilde{\Phi}_{f}^{\alpha\beta}(l,l')} = \frac{\partial F_{0}}{\partial \widetilde{\Phi}_{f}^{\alpha\beta}(l,l')} + \frac{\partial}{\partial \widetilde{\Phi}_{f}^{\alpha\beta}(l,l')} \Big\rangle H - \widetilde{H} \Big\langle_{0} = \frac{\hbar}{2MN} \sum_{\vec{k}} \frac{\sin^{2} \frac{k r_{ll'}}{2}}{\omega(\vec{k})} \operatorname{coth} \frac{\hbar \omega}{2\Theta} - \frac{y_{\alpha\beta}(l,l')}{4}.$$
(7)

In the non-equilibrium non-crystalline state, $\delta F \neq 0$. Expanding F into a power series in the system deviation from the equilibrium state and restricting ourselves to the first terms of the expansion, we have:

$$F = F_0 + \frac{1}{2} \sum_{i,j} a_{ij} \eta_i \eta_j + \frac{c}{3} \eta^3 + \frac{b}{4} \eta^4, \qquad (8)$$

where the following notations are used:

$$\begin{split} \eta &= \sigma - \sigma_{e} , \eta_{y} = \left\{ \eta_{y}^{\alpha} \right\}, \eta_{y}^{\alpha} = y_{\alpha} (l, l') - y_{\alpha} (l, l')_{e} , \ \eta_{\Phi} = \left\{ \eta_{\Phi}^{\alpha} \right\}, \eta_{\Phi}^{\alpha} = \widetilde{\Phi}^{\alpha \alpha} (l, l') - \widetilde{\Phi}^{\alpha \alpha} (l, l')_{e} , \\ a_{ij} &= \left(\frac{\partial^{2} F}{\partial \eta_{i} \partial \eta_{j}} \right)_{e} , \ c = \frac{1}{2} \left(\frac{\partial^{3} F}{\partial \eta^{3}} \right)_{e} \ , \ b = \frac{1}{6} \left(\frac{\partial^{4} F}{\partial \eta^{4}} \right)_{e} \end{split}$$

and the allowance was made for $\left(\frac{\partial F}{\partial \eta_i}\right)_e = 0$ (the subscript "e" corresponds to

the equilibrium state). Based on (8), we can write:

$$\frac{\partial F}{\partial \sigma} = a_{11}\eta + a_{12}\eta_y + a_{13}\eta_{\Phi} + c\eta^2 + b\eta^3,$$

$$\frac{\partial F}{\partial \eta_y} = a_{12}\eta + a_{22}\eta_y, \frac{\partial F}{\partial \eta_{\Phi}} = a_{13}\eta + a_{33}\eta_{\Phi}.$$
(9)

To describe the kinetics of the order parameter variation we shall use the Landau-Khalatnikov regression equation

$$\frac{\partial \eta_{i}}{\partial t} = -\gamma_{i} \left(\frac{\partial F}{\partial \eta_{i}} \right)$$

which allows one to obtain in relations (9) the following system of non-linear kinetic equations. According to the Prigogine principle of mode subordination, which holds for the highly non-equilibrium systems [2]: $\frac{\partial \eta_y}{\partial t} = 0$, $\frac{\partial \eta_{\Phi}}{\partial t} = 0$. This allows one to reduce the number of order parameters: $\eta_y = -\left(\frac{a_{12}}{a_{22}}\right)\eta$, $\eta_{\Phi} = -\left(\frac{a_{13}}{a_{33}}\right)\eta$ and rewrite (9) in the following form:

$$\gamma_1 \frac{\partial \eta}{\partial t} = -a_0 \eta - c \eta^2 - b \eta^3, a_0 = -\left(a_{11}^2 - \frac{a_{12}}{a_{22}^2} - \frac{a_{13}}{a_{33}^2}\right).$$
(10)

The expansion coefficients are the functions of the temperature, pressure and control parameter, i.e. the cooling rate is $a_0 = a_0(T,P,q)$. Since in the equilibrium state $\delta F = 0, \delta^2 F > 0$ and $\eta = 0$, then, to ensure this, $a_0(T,P,q)$ must be a positively defined matrix, i.e. $a_0(T,P,q) > 0$. In the

non-equilibrium state, $\delta F \neq 0$, $\eta \neq 0$. To provide the formation of the stationary states with a non-zero order parameter, it is essential that $a_0(T, P, q) < 0$. Thus, at the transition to the non-equilibrium state with the non-zero order parameter: $\eta = \frac{-2c \pm \sqrt{4c^2 - 4a_0b}}{6b}$, and the expansion coefficient is $a_0(T_c, P_c, q_c) = 0$. Here the following conditions must hold: $c(T_c, P_c, q_c) = 0, b(T_c, P_c, q_c) > 0$, since, with approaching the point T_c, P_c, q_c from the equilibrium state side, an even derivative should be positive. Thus, in the process of melt cooling and at the transition to the noncrystalline state, $a_0(T_c, P_c, q_c)$ can be approximated by:

$$a_{0}(T_{c}, P_{c}, q_{c}) = -\widetilde{a}_{0} \ln(\arctan[\widetilde{q}]), \widetilde{q} = \frac{q-q_{c}}{q_{c}}$$

which is transformed to $a_0(T_c, P_c, q_c) \approx -a_0 \tilde{q}$ as $\tilde{q} \ll 0$ [15].

Thus, having started from expressions (5)-(10), we arrive at the following self-consistent system of equations with respect to the

relative fraction of atoms in the liquid-like states, the mean-square atomic displacements $y_{\alpha\beta}(l,l')$ and the force constants $\widetilde{\Phi}_{f}^{\alpha\beta}(l,l')$:

$$F_{1}(\sigma) = 0, F_{2}(y) = 0, F_{3}(\Phi) = 0, \qquad (11)$$

$$F_{1}(\sigma) = \widetilde{T}_{2} - \widetilde{T}_{1} + 2\sigma (\widetilde{\Phi}_{2} - \widetilde{\Phi}_{1}) - 2\widetilde{\Phi}_{1} - 4\widetilde{I}(1 - \sigma)\sigma - 2\Theta N \ln \frac{\frac{g_{2}}{\sigma} - 1}{\frac{g_{1}}{1 - \sigma} - 1} + \widetilde{a}_{0}\widetilde{q}\eta - c\eta^{2} - b\eta^{3} - D_{\eta}\nabla^{2}\eta$$

$$F_{2}(y) = \frac{\hbar}{2MN} \sum_{k} \frac{\sin^{2} \frac{\vec{k}\vec{r}_{ll'}}{2}}{\omega(\vec{k})} \coth \frac{\hbar\omega}{2\Theta} - \frac{y_{\alpha\beta}(l, l')}{4} - \frac{\eta_{y}}{\tau_{y}}, \qquad (12)$$

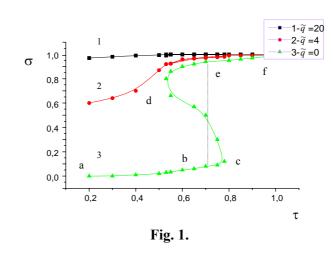
$$F_{3}(\Phi) = \frac{1}{4} (\sqrt[3]{\nabla}_{ll'}^{\alpha} \nabla_{ll'}^{\beta} \Phi_{r}(\vec{r}_{ll'}) / (\sigma_{r}^{2} - \widetilde{\Phi}_{r}^{\alpha\beta}(l, l')) - \frac{\eta_{\Phi}}{\tau_{\Phi}}.$$

This system (11), (12) allows the temperature properties of the system to be studied both at the equilibrium $(\eta = 0)$ and highly non-equilibrium $(\eta \neq 0)$ transformations.

III. Results and discussion

Consider the application of the above concepts to the studies of the stable and metastable states formation in the certain synergetics model. For the equilibrium transition "crystal – liquid" the ab area corresponds to the crystalline state, the ef area denotes the liquid and bc, de areas correspond to the metastable states, i.e. to the overheated crystal and the overcooled liquid, respectively (see fig. 1, τ is a reduced temperature).

Let us analyze the temperature behaviour of the solutions of the system of equations highly non-equilibrium (11), (12) in conditions, i.e. at $\tilde{q} \rightarrow \tilde{q}_{c}$. The temperature dependence of the fraction of atoms in the "soft" states have two specific domains (Figs. 1, 2). First of them corresponds to the system cooling at $\tilde{q} < \tilde{q}_c$, when the transition to the solid state occurs with the spasmodic increase in the elastic constants and the decrease in the fraction of atoms in the "soft" states. Note that the value of the leap decreases with increasing \tilde{q} . The second domain observed at $\tilde{q} > \tilde{q}_c$ is characterized by the continuous anomalous increase, the development of a shear rigidity of the structure and the elastic constants, as well the reduction of (see continuous Fig. 2).



Consequently, the curve at $\tilde{q} = \tilde{q}_c$ demarcates the region of the existence of metastable (i.e. the overcooled liquid) and highly non-(non-crystalline) equilibrium condensed systems. It defines the lower limit of the possible reduction of the mean-square atom displacements and the fraction of atoms in the "soft" states in the temperature range above τ_0 at $\tilde{q} < \tilde{q}_c$ (τ_0 is the temperature of branche f-F on Fig. 2). The synthesis, temperature τ_c in the curve \tilde{q}_c , at which metastable states are degenerated, is the temperature of coexistence of three states at a preset pressure, i.e. crystalline, liquid and $\left(rac{\partial^2 \sigma}{\partial au^2}
ight)$ =0 (point S, non-crystalline states: Fig. 2). The studies of the peculiarities in the

behaviour of the thermodynamic properties of the system in the vicinity of $\{\tau_c, q_c\}$ are of doubtless interest. Note that the correlated reduction of the intensity of atom vibrations and the rise of the force constants at the transition from the metastable overcooled melt to the non-crystalline solid gives evidence for the occurrence of the collective macroscopic processes at the microstructural rearrangement level at which the noncrystalline structure with the inner order parameter is formed depending on the control parameter values \tilde{q} . The order parameter used determines the degree of system deviation from the equilibrium state and correlates, in particular, with the fluctuations of system deviation from the Gaussian distribution. The non-equilibrium stationary

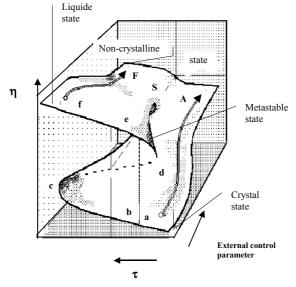


Fig. 2.

structure produced at $\tilde{q} > \tilde{q}_c$ results from the instability of the ground thermodynamically non-equilibrium state and arises through the self-consistent enhancement of fluctuations (the mean-square atomic displacements and the fraction of atoms in the "soft" states) which reach the macroscopic level and make a new structure stable. That structure is a dissipative one.

Indeed, when the melt is cooled down under highly non-equilibrium conditions, the part of the energy of the system, which is related to the thermal motion of particles, is transformed into the macroscopically organized motion due to the spatial-temporal correlations at the macroscopic scale. This motion is transformed into the complicated spatial organization of the system. Such a method of organization just constitutes the dissipative structure. The dissipative structure formation in non-crystalline solids under cooling is related to the self-consistent creation of the "soft"-state domains and results from the temperature behaviour of the structures under evolution towards those changes, which may promote the minimum energy dissipation and facilitate the technological process.

Acknowledgment

This work was supported by an Ukrainian-Hungarian cooperation grant № 2M/182-2000.

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Процеси самоорганізації та формування дисипативних структур в некристалічних матеріалах

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Формування дисипативних структур в некристалічних матеріалах пов'язано з явищем самоорганізації. В рамках даного підходу некристалічний стан розглядається як результат попередньої самоорганізації, який відповідає мінімуму дисипації енергії при даних умовах одержання.