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COMPOSITE MATERIALS WITH NEGATIVE POISSON COEFFICIENT

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A theoretical approach is suggested for the creation of composite materials based on disperse filler-filled polymers with a negative Poisson coefficient. Necessary and sufficient conditions are formulated for it.

Production of polymer systems with a negative Poisson coefficient (γ) is of particular theoretical and applied interest [1].

In the experiments use was made of thermoplastic polyurethanes (TPU) synthesized on the basis of 4,4-diphenylmethanediisocyanate, 1,4-butanediol, and polyoxytetramethylenglycol with MM = 1500. Highly dispersed Fe, Mo, and W powders with a particle size of 0.3-1 μm were used as fillers. The specimens were prepared in the $T-p$ regime [2]. At a frequency of 0.4 MHz, the specific-heat increment (ΔC) and the rates of longitudinal (ν_{long}) and shear ν_{sh} deformations were determined following the method described in [3].

Analysis of the temperature and concentration dependences of the specific heat of the systems has shown that as the content of the fillers increases, the portion of TPU macromolecules that go to the boundary layer increases [2]. It should be noted that an important characteristic of the microstructure of the composites is formation of spatial coagulation structures in two cases of the content of the filler in the system (φ): (1) when it is lower than the critical content (φ_{cr}) and (2) equal to or exceeding the critical content.

At $\varphi < \varphi_{\mathrm{cr}}$, in the absence of segregation, particles of a highly dispersed filler are not at the nodes of the regular structure but occupy random positions in space. Initially, such a heterogeneous system can be simulated by a quasi-one-dimensional lattice with a random distance between particles of the filler [4]. In this case there is no cooperative motion of the particles, since the quasi-elastic component of the interaction force does not appear at the phase interface. The particles of the filler that are in the polymer matrix experience individualized action under the total action of friction forces or the medium's resistance forces. The rate of their Brownian motion is determined by the temperature of the thermostat, the filler particle size, and the viscosity of the medium. An external force will make the particles move as colloid particles in a high-viscosity medium, irrespective of the motion of their neighbors [5].

As the content of the highly dispersed filler in the system increases, a larger and larger amount of the polymer matrix is transformed to the state of the boundary layer, which "displaces" the polymer in space. At $\varphi \geq \varphi_{\mathrm{cr}}$, a three-component heterogeneous system degenerates to a two-component system of the filler-boundary layer type [6]. In the case of $\varphi = \varphi_{\mathrm{cr}}$, spatially ordered systems appear, since particle motion depends on the motion of their neighbors. Such a linear system is a one-dimensional crystal with the period [7, 8]

$$l = d\left[\left(\frac{\pi}{6}\,\frac{\rho_{\mathrm{f}}/\rho_{\mathrm{p}} + \varphi_{\mathrm{cr}}}{1.91\varphi_{\mathrm{cr}}}\right)^{1/3} - 1\right], \qquad (1)$$

where d is the filler particle diameter and ρ_{f} and ρ_{p} are the densities of the filler and polymer, respectively.

In the absence of a spatial coagulation structure of a heterogeneous system, particles of the filler form a simple cubic lattice as a possible type of structure of a body with a minimum volume. Its nodes are imbedded in the polymer matrix, which is in the boundary-layer state. The period of this superlattice [4] is l and the internal

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field is materialized by a polymer "binder" effecting interaction between particles of the filler. The polymer "binder" will be simultated by elastic bonds with constant β in the longitudinal direction and κ in the transverse direction.

By extrapolation of the relation $\Delta C_f/\Delta C \to 0$, where ΔC_f and ΔC are the values of the specific-heat increment for the filled system and the initial polymer, the critical contents φ_{cr} of Fe, Mo, and W in the system are found to be 43, 48, and 52 vol.%, respectively.

Calculations by Eq. (1) have shown that l is commensurable with the filler particle size, and for the system considered it varies in the range of $(0.46-0.80)d$ [9].

We consider a metal-filled polymer matrix based on a TPU. Unlike atoms in the interior, surface metal atoms are partly hydrated by the presence of hydroxyl groups. Oxidation, for example, of Fe, follows the scheme $Fe \to FeO \to Fe_3O_4 \to 0$. However, in this case on the surface of a highly dispersed filler there exists 1–2% coordination-unsaturated metal atoms, and it is these atoms that start to interact with the TPU [2]. In order to obtain an equation of state for this condensed system, the interaction potential of its atoms will be expressed as a sum of two terms: $\varphi_1(r)$ and $\varphi_2(r)$ [10]. The first term $\varphi_1(r)$ is assumed to be equal to the Lennard–Jones potential, which characterizes interatomic interaction at $T = 0$. In order to determine the contribution of the elastic component of atomic interactions to the specific internal energy of the system, the potential interaction energy will be summed over all possible parameters of the atoms that are paired with the atom considered, and the result will be multiplied by the number of atoms. The second term $\varphi_2(r)$ of the interaction potential is a function of T and is related to the pressure that produces a flow of carriers emitted by an atom to the other atoms in the body.

We consider the flow of carriers that are emitted by a surface atom of a highly dispersed metal filler as an active center of interaction with a TPU. This atom α exerts pressure $f_{\alpha\beta}$ on atom β (TPU), which is proportional (γ) to the absorption cross-section of the atom $\beta(S_\alpha)$, irrespective of the type of carrier and the energy (E_α), and decreases as the distance (r) from atom α increases, i.e., $f_{\alpha\beta} = \gamma S_\alpha k E_\alpha / r^m$, where k and m are positive constants and $m > 2$ [10]. In this case, the component of the interatomic potential $\varphi_2(r)$ is defined by

$$\varphi_2(r) = -\int f_{\alpha\beta}(r)\,dr = -\int (\gamma S_\alpha k E_\alpha / r^m)\,dr = BE\left(\frac{a}{r}\right)^{m_3},$$

where $\gamma S_\alpha k(-m+1) = B$; $E_\alpha = E$; $m_3 = -m + 1$, and a are positive constants.

Then, the potential of interaction of this atom with any other atom in the polymer matrix can be expressed as the sum

$$\varphi(r) = 4D\left[\left(\frac{a}{r}\right)^{12} - \left(\frac{a}{r}\right)^{6}\right] + BE\left(\frac{a}{r}\right)^{m_3}, \tag{2}$$

where D and a are constants with dimensions of energy and length, respectively. The length a is the equilibrium distance between the atoms, and at $a = r$ we have $\varphi(a) = 0$, when $T = 0$ ($E = 0$ at $T = 0$).

Proceeding from relation (2), we find the force acting between these atoms and holding them in a certain position relative to each other:

$$f(r) = -\frac{d\varphi(r)}{dr} = 24D\left(2\frac{a^{12}}{r^{13}} - \frac{a^6}{r^7}\right) + \frac{BEm_3}{a}\left(\frac{a}{r}\right)^{m_3+1}. \tag{3}$$

It should be noted that while in Eq. (2) the exponent 6 at the term that describes attraction is theoretically reasonable and consistent with quantum-mechanics calculations, the exponent 12 at the term that describes collision is not justified so strictly. Therefore, in what follows $m_1 \geq 12$ and $m_2 = 6$ will be assumed in most general form.

The presence of interaction force (3) between atoms of the filler and the polymer matrix promotes further structurization of the system, restricting the mobility of polymer chains, which changes the conditions of the relaxation processes. This is equivalent to the appearance of additional internal pressure, establishes a new quasi-equilibrium state of the composite. For determination of its value it is assumed that the atoms of the polymer matrix are uniformly distributed, with density n_i, over concentric spherical surfaces located at equal distances from the surface of the highly dispersed filler. On the surface z_i a unit area is isolated; then the resultant of the forces of interaction of the atom with all the other atoms in the polymer matrix is defined as follows:

$$F = \sum_i F_{ai} = \sum_i \int_0^\infty dF_i 2\pi n_i \left[A \left(\frac{a^{m_1}}{z_{ai}^{m_1-1}} - \frac{a^{m_2}}{z_{ai}^{m_2-1}} \right) + B\bar{E} \frac{a^{m_3}}{z_{ai}^{m_3-1}} \right] \frac{z_{ki}}{z_{ai}}, \tag{4}$$

where $\bar{E} = (1/n_i) \sum_{\alpha=1}^{n_i} E_\alpha$; $z_{ki} = |z_{\alpha i}| = |z_\alpha - z_i|$.

In view of the fact that at $\varphi \geq \varphi_{\text{cr}}$ a heterogeneous polymer system is a set of macrolattices, we choose one of them with period l in the composite and consider the total force exerted on it. The unknown total force is determined as the sum of only those forces that are exerted on the volume considered by parts of the body surrounding it. Since these forces act on the macrolattice through its surface, then, apart from interatomic interaction forces (4), surface forces F_s can act on particles of the body in the direction of the axis z. If a uniform stress G acts in the direction of the normal to the surface, the average surface force per atom in the external layer is $F_1 = G/n_0$. Accordingly, the equilibrium equation for an arbitrary atom in the external layer can be expressed as

$$F + F_s = m \frac{d^2 z}{dt^2}. \tag{5}$$

In the equilibrium state $d^2z/dt^2 = 0$, with allowance for the fact that the force exerted on the entire surface of the body by internal stresses is $\oint G dS$, per atom of the macrolattice, we define $G = (F/h^2) n_0$. Then, in accordance with (4) and (5), the average surface force is found from equilibrium for atoms in the external layer ($z_\alpha = z_0 = 0$) in the form

$$F_s = \frac{G}{n_0} = -2\pi n_0 \left[A \left(\frac{a^{m_1}}{h^{m_1-1}} \sum_{i=1}^\infty \frac{1}{i^{m_1-1}} - \frac{a^{m_2}}{h^{m_2-1}} \sum_{i=1}^\infty \frac{1}{i^{m_2-1}} \right) + BE \frac{a^{m_3}}{h^{m_3-1}} \sum_{i=1}^\infty \frac{1}{i^{m_3-1}} \right], \tag{6}$$

where $h = h_i = z_{i+1} - z_i$ is the distance between adjacent layers of the macrolattice parallel to the surface on which the external forces act. In view of the fact that the internal stresses are caused by molecular forces that have an insignificant effective range, according to (6), calculation of G is reduced to consideration of h (as well as the rapid convergence of $\sum_{i=1}^{\infty} (1/i^{m_1-1})$ of the order of magnitude of the interatomic distance. Consequently, relation (6) is similar to (3) in its form. Since with deformation of the system, a pressure (p) equal in magnitude and directed everywhere along the normal to the surface of the macrolattice acts on every unit volume, with allowance for the fact that $p = F_s n_0 = F_s/h^2$ and $h^3 = v_1 = vM/N_0$, where v_1 is the volume per atom and v is the specific volume of the compound, Eq. (6) can be written as

$$p = A_1 \left[\left(\frac{v_0}{v} \right)^{u_1} - \left(\frac{v_0}{v} \right)^{u_2} \right] + A_2 \bar{E} \left(\frac{v_0}{v} \right)^{u_3}, \tag{7}$$

where $v_0 = a^3$; $v = r^3$; A_1, A_2, u_1, u_2, u_3 are positive constants according to Eqs. (3) and (6).

We consider the case of deformation of the composite accompanied by changes in volume without changes in shape.

In the case of such uniform deformation of the composite, with allowance for the fact that on the surface of the microlattice $G_{zz} n_i = p$, i.e., $p = G_{zz}$, and the relative change in the specific volume v is slight, in the Taylor series of $(v_0/v)^{u_i}$ we can restrict ourselves to terms of the second order of smallness. Then

$$\left(\frac{v_0}{v} \right)^{u_i} = \left(1 + \frac{\Delta v_0}{v} \right)^{-u_i} \simeq 1 - u_i \frac{\Delta v_0}{v}; \quad \Delta v = v - v_0, \quad i = 1, 2, 3, \ldots,$$

and at $T = 0$ relation (7) becomes

$$\frac{\Delta\nu_0}{\nu} = \frac{G}{\kappa}, \tag{8}$$

where $\kappa = A_1(u_1 - u_2)/(1 + u_3\Delta\nu_0/\nu) \simeq A(u_1 - u_2)$.

Using relation (3), we determine the maximum value of the force of interaction between atoms of the polymer and highly dispersed filler at the phase interface, with the extremum conditions $df(r)/dr = 0$ at $T = 0$ ($E = 0$). Then,

$$\frac{df(r)}{dr} = 24D\left(-26\frac{a^{12}}{r^{14}} + 7\frac{a^6}{r^8}\right). \tag{9}$$

With the distance between atoms of the polymer and highly dispersed filler $f(r) = F_{\max}$ denoted by $r = r_{\max}$, it is found that $r_{\max} = 1.245a = ba$. Consequently, in deformation of the system the maximum force of interaction between atoms

$$F_{\max} = 24D\left(2\frac{a^{12}}{r_{\max}^{13}} - \frac{a^6}{r_{\max}^{7}}\right) = \frac{24D}{b^7 a}\left(\frac{2}{b^6} - 1\right). \tag{10}$$

Since $2/b^6 - 1 < 0$ is the attraction force, the disruptive force is defined as

$$F_{\rm dis} = \frac{24D}{b^7 a}\left(1 - \frac{2}{b^6}\right). \tag{11}$$

Since in (6) the function F_s decreases quite rapidly as i increases at constant p and T, the density of macroscopic bodies is independent of their size. Then, the relative change in the specific volume of the system is found in terms of the structural characteristics of the macrolattice with allowance for its maximum elastic deformation as $(\Delta V/V_0) \simeq 3l^2 \cdot 1.245a/l^3 = 3 \cdot 1.245a/l$. In this case from relation (7) it is possible to determine the average range of internal stresses that must be created by an external force in a heterogeneous polymer system in order that $-1 < \gamma < 0$:

$$\frac{1.245a}{l}E < G < \frac{3 \cdot 1.245a}{l}E. \tag{12}$$

In the case of adiabatic deformations of the system, the ordinary (isothermal) quantities E and G should be replaced by their adiabatic values. It should be noted that in order to obtain the equation of elastic medium, we consider the interaction of its adjacent elements with masses M_A and M_B. Assuming that they form a linear chain of the type $ABAB\ldots$, [11, 12], for longitudinal vibration, the solution of a secular equation of the type

$$\left|\frac{M}{M_A M_B}\Phi_{A,B} - M\omega^2\delta_{A,B}\right| = 0, \tag{13}$$

where $\Phi_{A,B}$ is the potential of interaction between elements A and B, has the form

$$\omega(k) = \left[\frac{\beta}{M_A M_B}\left[M_A + M_B \pm \left(M_A^2 + M_B^2 + 2M_A M_B \cos(dk)\right)^{1/2}\right]\right]^{1/2}. \tag{14}$$

Assuming that the lattices A and B vibrate, on the whole, relative to each other ($\cos(dk) = 1$) and $\omega_{\max}(k) = \omega_{\rm D}^{\rm long}$, we obtain

$$\beta = \frac{(\omega_{\rm D}^{\rm l})^2 M_A M_B}{2(M_A + M_B)}, \tag{15}$$

where $\omega_{\mathrm{D}}^{\mathrm{long}}$ is the longitudinal characteristic frequency.

Since transverse vibrations are caused by flexibility of the chain and effected by deformation of valence angles, the force that arises in this case is governed by Hooke's law. Then,

$$\omega(k) = \left(\frac{4\kappa}{M}\right)^{1/2} \sin^2 \frac{kd}{2}, \tag{16}$$

where $\omega(k)$ is the transverse characteristic frequency.

Assuming that $\omega_{\max}(k) = \omega_{\mathrm{D}}^{\mathrm{sh}}$, we find

$$\kappa = \frac{(\omega_{\mathrm{D}}^{\mathrm{sh}})^2 M}{4}. \tag{17}$$

The characteristic frequency spectrum ($\omega_{\mathrm{D}}^{\mathrm{long}}$, $\omega_{\mathrm{D}}^{\mathrm{sh}}$) was calculated using the rates of longitudinal ν_{long} and shear ν_{sh} deformations by the relations

$$\omega_{\mathrm{D}}^{\mathrm{long}} = \left(\frac{6\pi^2 N}{V}\right)^{1/3} \nu_{\mathrm{long}}; \quad \omega_{\mathrm{D}}^{\mathrm{sh}} = \left(\frac{6\pi^2 N}{V}\right)^{1/3} \nu_{\mathrm{sh}}. \tag{18}$$

If it is assumed now that in a cylindrical body with cross-sectional area S under deformation, motion occurs in the form of small elastic vibrations or waves, then their average power can be expressed in the form

$$\langle p_{\mathrm{long}} \rangle = \langle F_{\mathrm{long}} \rangle \langle \nu_{\mathrm{long}} \rangle, \tag{19}$$

$$\langle p_{\mathrm{sh}} \rangle = \langle F_{\mathrm{sh}} \rangle \langle \nu_{\mathrm{sh}} \rangle. \tag{20}$$

Accordingly, the intensity is expressed as

$$\langle I_{\mathrm{long}} \rangle = \frac{\langle p_{\mathrm{long}} \rangle}{S} = \frac{\langle F_{\mathrm{long}} \rangle \langle \nu_{\mathrm{long}} \rangle}{S}, \tag{21}$$

$$\langle I_{\mathrm{sh}} \rangle = \frac{\langle p_{\mathrm{sh}} \rangle}{S} = \frac{\langle F_{\mathrm{sh}} \rangle \langle \nu_{\mathrm{sh}} \rangle}{S}. \tag{22}$$

If it is assumed that the velocity of, for example, ultrasound, is the same in both the longitudinal (OY) and transverse (OX) directions (this can be achieved by carrying out experimental studies), at $\langle I_{\mathrm{long}} \rangle = \langle I_{\mathrm{sh}} \rangle = \langle I \rangle$, equations (21) and (22) can be expressed as

$$\langle I \rangle = \frac{\langle F_{\mathrm{long}} \rangle \langle \nu_{\mathrm{long}} \rangle}{S}, \tag{23}$$

$$\langle I \rangle = \frac{\langle F_{\mathrm{sh}} \rangle \langle \nu_{\mathrm{sh}} \rangle}{S}. \tag{24}$$

Taking into consideration that

$$\langle F_{\mathrm{long}} \rangle = \frac{\beta y_0}{2}, \tag{25}$$

$$\langle F_{\mathrm{sh}} \rangle = \frac{\kappa x_0}{2}, \tag{26}$$

where x_0 and y_0 are displacements of adjacent atoms from the equilibrium position in the corresponding directions.

With the assumption that the wavelength is much longer than the distance between adjacent atoms, in view of (15) and (16) at $M_A \simeq M_B$, according to (25) and (26) Eqs. (23) and (24) are expressed in the form:

$$\langle I \rangle = \left(\frac{\omega_{\mathrm{D}}^{\mathrm{long}}}{2} \right)^2 \langle \rho \rangle \langle v_{\mathrm{long}} \rangle y_0^2 , \tag{27}$$

$$\langle I \rangle = \left(\frac{\omega_{\mathrm{D}}^{\mathrm{sh}}}{2} \right)^2 \langle \rho \rangle \langle v_{\mathrm{sh}} \rangle x_0^2 , \tag{28}$$

Hence, finally

$$y_0^2 = - \frac{\langle I \rangle}{\left(\frac{\omega_{\mathrm{D}}^{\mathrm{long}}}{2} \right)^2 \langle \rho \rangle \langle v_{\mathrm{long}} \rangle} , \tag{29}$$

$$x_0^2 = \frac{\langle I \rangle}{\left(\frac{\omega_{\mathrm{D}}^{\mathrm{sh}}}{2} \right)^2 \langle \rho \rangle \langle v_{\mathrm{sh}} \rangle} . \tag{30}$$

We introduce the notation

$$a_1 = \frac{\langle I \rangle}{\left(\frac{\omega_{\mathrm{D}}^{\mathrm{long}}}{2} \right)^2 \langle \rho \rangle} , \tag{31}$$

$$a_2 = \frac{\langle I \rangle}{\left(\frac{\omega_{\mathrm{D}}^{\mathrm{sh}}}{2} \right)^2 \langle \rho \rangle} . \tag{32}$$

With allowance for (31) and (32), relations (29) and (30) can be expressed as

$$y_0^2 = \frac{a_1}{\langle v_{\mathrm{long}} \rangle} , \tag{33}$$

$$x_0^2 = \frac{a_2}{\langle v_{\mathrm{sh}} \rangle} . \tag{34}$$

We transform (33) and (34) to

$$\frac{y_0^2}{2 \frac{a_1}{\langle v_{\mathrm{long}} \rangle}} = \frac{1}{2} , \tag{35}$$

$$\frac{x_0^2}{2 \frac{a_2}{\langle v_{\mathrm{sh}} \rangle}} = \frac{1}{2} . \tag{36}$$

TABLE 1. Dependence of Structural-Mechanical Parameters on the Composite Type

Structural-mechanical parameters of the system	Composite type		
	TPU + W	TPU + Mo	TPU + Fe
φ_{cr}, vol.%	48	43	52
$\omega_{\mathrm{D}}^{\mathrm{long}} \cdot 10^{-13}$, Hz	1.421	1.565	1.468
$\omega_{\mathrm{D}}^{\mathrm{sh}} \cdot 10^{-13}$, Hz	1.237	1.253	1.468
$\omega_{\mathrm{D}} \cdot 10^{-13}$, Hz	1.288	1.329	1.239
$\Theta_{\mathrm{D}}^{\mathrm{long}}$, K	109	120	112
$\Theta_{\mathrm{D}}^{\mathrm{sh}}$, K	95	96	90
Θ_{D}, K	98	102	95
a, Å	2.08	1.84	2.06
l, μm	0.71	0.69	0.63
β, N/m	1.638	1.986	1.747
κ, N/m	0.443	0.461	0.402
γ	−0.397	−0.365	−0.236

Then, we obtain

$$\frac{y_0^2}{\left(\sqrt{\left(2\frac{a_1}{\langle\nu_{\mathrm{long}}\rangle}\right)}\right)^2} + \frac{x_0^2}{\left(\sqrt{\left(2\frac{a_2}{\langle\nu_{\mathrm{sh}}\rangle}\right)}\right)^2} = 1\,. \tag{37}$$

Consequently, in systems with $\gamma < 0$, the deformation process of elastic vibrations propagates in the form of an expanding ellipsoid of revolution. This can be observed by the method of holographic interference. Experiments carried out with nickel alloy single crystals [13] have shown definitely that in the case of $\gamma < 0$, the interference processes actually have the form of a set of ellipsoids. In this case, the average γ is determined as $-a^2/b^2$, where a and b are semiaxes of the ellipses in directions x and y, respectively. It follows from relation (37) that $\gamma = -(\nu_{\mathrm{sh}}/\nu_{\mathrm{long}}) \cdot (\omega_{\mathrm{D}}^{\mathrm{sh}}/\omega_{\mathrm{D}}^{\mathrm{sh}})^2$. In this case for the systems TPU+Mo, TPU+Fe, and TPU+W, γ is equal to −0.42, −0.40, and −0.25, respectively, which agrees satisfactorily with calculations (Table 1).

From relations (12) and (13) it is possible to determine the internal stresses in the case of a dynamic load on the body for the case of $-1 < \gamma < 0$. With allowance for adiabatic deformation and the values of $\nu_{\mathrm{long}} = [(\lambda + 2\mu)/\rho]^{1/2}$ and $\nu_{\mathrm{sh}} = (\mu/\rho)^{1/2}$ from [14], where λ and μ are Lame's constants, using relations (15)-(17), we find the analytical expression

$$E = \frac{4\rho}{M}\left(\frac{V}{6\pi^2 N}\right)^{2/3}(\beta + 2\kappa)\,.$$

In this case, the average range of internal stresses that should be created by an external force field in a heterogeneous system in order that it have $-1 < \gamma < 0$ is found, according to (12), as

$$\frac{1.245a\cdot 4\rho}{lM}\left(\frac{V}{6\pi^2 N}\right)^{2/3}(\beta + 2\kappa) < \sigma < \frac{3\cdot 1.245a\cdot 4\rho}{lM}\left(\frac{V}{6\pi^2 N}\right)^{2/3}(\beta + 2\kappa)\,. \tag{38}$$

The calculations (Table 1) show that for the TPU systems considered a negative Poisson coefficient is observed if they have internal stresses in the range of $0.97\cdot 10^6$ Pa $< \sigma <$ $7.11\cdot 10^6$ Pa.

Thus, necessary and sufficient conditions are established under which structural modification of polymer systems is possible at $\varphi = \varphi_{cr}$ in order that $-1 < \gamma < 0$.

NOTATION

γ, Poisson coefficient; β, κ, elasticity constants; $f(r)$, force of interaction; $\varphi(r)$, interatomic potential; σ, internal stress.

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