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Temperature dependence effect of viscosity on ultrathin lubricant film melting*

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We study the melting of an ultrathin lubricant film under friction between atomically flat surfaces at temperature dependencies of viscosity described by Vogel-Fulcher relationship and by power expression, which are observed experimentally. It is shown that the critical temperature exists in both cases the exceeding of which leads to the melting of lubricant and, as a result, the sliding mode of friction sets in. The values of characteristic parameters of lubricant are defined, which are needed for friction reduction. In the systems, where the Vogel-Fulcher dependence is fulfilled, it is possible to choose the parameters at which the melting of lubricant takes place even at zero temperature of friction surfaces. The deformational defect of the shear modulus is taken into account in describing the lubricant melting according to the mechanism of the first-order transition.

Key words: *viscoelastic medium, boundary friction, shear stress and strain*

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1. Introduction

The problems of sliding friction continue to attract a considerable attention [1–3] due to the phenomenon being vastly used in the engineering of smooth solid surfaces, divided by thin films of lubricant. In works [4–10] the boundary friction is described which is realized in the case of ultrathin films with thickness less than four diameters of molecules, at smooth or rough surfaces, high loads and small shear rates. It is known that shear viscosity of lubricant layer depends on a temperature [11–14]. Thus, at the derivation of basic equations describing the process of friction, it is necessary to define the explicit form of temperature dependence of viscosity. In the previous works [4–10] the simplest dependence of viscosity on temperature was used. However, actually, it takes different forms for specific systems. According to the experimental results this function is usually fixed by either Vogel-Fulcher relationship or by power dependence [11–14]. In particular, the Vogel-Fulcher empirical formula is explained within the framework of theories of free volume and excess configuration entropy [11]. These two ideas are realized in the Ising-Nakano microscopic model. The introduction of the temperature-dependent effective field makes it possible to take into account the conservation of total free volume at the defined external conditions [15]. At the presence of the frozen disorder of interatomic connections, the glass transition is presented as rheological transition.

The purpose of this work is to take into account the indicated dependencies in the earlier proposed rheological model in describing the viscoelastic lubricant [4]. It allows us to spread the developed approach to a wider class of the systems. As a result, the expressions for the melting temperature of the lubricant are found. It is shown that systems, in which the temperature dependence of the viscosity is fixed by the Vogel-Fulcher relationship, are preferable in forming the conditions of friction reduction. This is caused by the possibility of choosing the optimal parameters providing the liquid-like structure of lubricant at an arbitrary temperature of the sheared surfaces. In the case of the power dependence for the viscosity being realized, the critical temperature is

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limited by the minimal non-zero value that is independent of elastic characteristics of lubricating material.

2. Basic equations

In the previous work [4] based on the rheological description of viscoelastic medium possessing the heat conductivity, a system of kinetic equations has been obtained, which determine the mutually coordinated behaviour of the shear stress σ and strain ε, and the temperature T in an ultrathin film of lubricant during friction between atomically flat mica surfaces. Let us write down these equations introducing the units of measurement for variables σ, ε, T:

$$\sigma_s = \left(\frac{\rho c_v \eta_0 T_c}{\tau_T}\right)^{1/2}, \qquad \varepsilon_s = \frac{\sigma_s}{G_0} = \left(\frac{\tau_\varepsilon}{\tau_T}\right)^{1/2}\left(\frac{\rho c_v T_c \tau_\varepsilon}{\eta_0}\right)^{1/2}, \qquad T_c\,, \tag{1}$$

where T_c is the characteristic temperature, $\tau_T \equiv \rho l^2 c_v/\kappa$ is the time of heat conductivity, κ is the coefficient of heat conductivity, l is the length of heat conductivity, $\eta_0 \equiv \eta(T = 2T_c)$ is the characteristic value of viscosity, ρ and c_v are the density and the specific heat capacity of lubricant, $G_0 \equiv \eta_0/\tau_\varepsilon$, τ_ε is the strain relaxation time:

$$\tau_\sigma \dot{\sigma} = -\sigma + g\varepsilon, \tag{2}$$

$$\tau_\varepsilon \dot{\varepsilon} = -\varepsilon + (T-1)\sigma, \tag{3}$$

$$\tau_T \dot{T} = (T_e - T) - \sigma\varepsilon + \sigma^2. \tag{4}$$

Here the stress relaxation time τ_σ, the temperature T_e of atomically flat mica friction surfaces, and the constant $g = G/G_0$ are introduced, where G is the lubricant shear modulus. Replacement of ε/τ_σ by $\partial\varepsilon/\partial t$ reduces equation (2) to the Maxwell-type equation for viscoelastic matter approximation that is widely used in the theory of boundary friction [1]. The relaxation behaviour of viscoelastic lubricant during the process of friction is also described by Kelvin-Voigt equation (3) [4,16]. Equations (2) and (3) jointly represent the new rheological model. It is worth noting that rheological properties of lubricant films are investigated experimentally in order to construct a phase diagram [3]. Equation (4) represents the heat conductivity expression, that describes the heat transfer from the friction surfaces to the layer of lubricant, the effect of dissipative heating of a viscous liquid flowing under the action of the stress, and the reversible mechanic-and-caloric effect in linear approximation. Equations (2)–(4) formally coincide with the synergetic Lorentz system [17,18], where the shear stress acts as the order parameter, the conjugate field is reduced to the shear strain, and the temperature is the control parameter. As is known this system can be used in describing the thermodynamic phase and the kinetic transitions.

In reference [4] the melting of ultrathin lubricant film under friction between atomically flat mica surfaces is represented as a result of the action of spontaneously appearing shear stress leading to the plastic flow. This is caused by the heating of friction surfaces above the critical value $T_{c0} = 1 + g^{-1}$. Thus, according to this approach the studied solid-like–liquid-like transition of lubricant film occurs due to both thermodynamic and shear melting. The initial reason for this self-organizing process is the positive feedback of T and σ on ε (see equation (3)) conditioned by the temperature dependence of the shear viscosity leading to its divergence. On the other hand, the negative feedback of σ and ε on T in equation (4) plays an important role since it ensures the system stability.

According to this approach the lubricant represents a strongly viscous liquid that can behave similar to the solid. It has a high effective viscosity and still exhibits a yield stress [3,16]. Its solid-like state corresponds to the shear stress $\sigma = 0$ because equation (2) falls out of consideration. Equation (3), containing the viscous stress, reduces to the Debye law that presents a rapid relaxation of the shear strain during the microscopic time $\tau_\varepsilon \approx a/c \sim 10^{-12}$ s, where $a \sim 1$ nm is the lattice constant or the intermolecular distance and $c \sim 10^3$ m/s is the sound velocity. At that the heat conductivity equation (4) takes the form of the simplest expression for temperature relaxation that does not contain the terms representing the dissipative heating and the mechanic-and-caloric effect of a viscous liquid.

Equation (3) describes the flow of lubricant with velocity $V = l\partial\varepsilon/\partial t$ due to the action of the appearing viscous shear stress. Moreover, according to reference [19] in the absence of shear deformations the temperature mean-square displacement is defined by equality $\langle u^2\rangle = T/Ga$. The average shear displacement is found from the relationship $\langle u^2\rangle = \sigma^2 a^2/G^2$. The total mean-square displacement represents the sum of these expressions provided that the thermal fluctuations and the stress are independent. The above implies that the transition of lubricant from solid-like to fluid-like state is induced both by heating and under the effect of stress generated by solid surfaces at friction. This agrees with the examination of solid state instability within the framework of shear and dynamic disorder-driven melting representation in the absence of thermal fluctuations [19]. It is shown [20] that the plastic flow of lubricant layer is realized at the presence of elastic stress. The action of shear stress causes the reduction of shear modulus of lubricating material [21]. Consequently, the friction force decreases with the velocity increase at the contact $V = l\partial\varepsilon/\partial t$ because the latter leads to the growth of the shear stress σ according to the Maxwell stress – strain ε relationship: $\partial\sigma/\partial t = -\sigma/\tau_\sigma + G\partial\varepsilon/\partial t$. It is assumed that the film becomes more liquid-like and the friction force decreases with the temperature growth due to activation energy barrier decreasing to molecular hops.

At derivation of equation (3) the simplest temperature dependence of viscosity η was defined in relation to the critical temperature [12]:

$$\eta(T) = \frac{\eta_0}{T/T_\mathrm{c} - 1}\,. \tag{5}$$

The purpose of the proposed work is to describe the lubricant melting process at different dependencies $\eta(T)$. According to the experimental data the function $\eta(T)$ is defined either by power expression

$$\eta(T) = \frac{\eta_0}{(T/T_\mathrm{c} - 1)^\gamma}\,, \tag{6}$$

which generalizes the formula (5) , or by Vogel-Fulcher relationship

$$\eta(T) = \eta_0' \exp\frac{B'}{(T - T_\mathrm{c})^\alpha}\,, \tag{7}$$

where γ, η_0', B', and α are the positive constants [11–14].

3. Vogel-Fulcher relationship

Using the relationship (7) and introducing the dimensionless constants $A \equiv \tau_\varepsilon G_0/\eta_0'$, $B \equiv B' T_\mathrm{c}^{-\alpha}$ the equations (2), (4) are not changed, and (3) assumes the form:

$$\tau_\varepsilon \dot{\varepsilon} = -\varepsilon + A\sigma \exp\left[-\frac{B}{(T-1)^\alpha}\right]. \tag{8}$$

In order to analyse the modified system we use adiabatic approximation within the framework of which the characteristic time scales satisfy the inequality [4,5]:

$$\tau_\sigma \gg \tau_\varepsilon, \tau_T\,. \tag{9}$$

It means that in the course of evolution the strain $\varepsilon(t)$ and the temperature $T(t)$ follow the change of the stress $\sigma(t)$ [17,18]. Then, the left-hand sides of equations (8) and (4) can be set equal to zero. As a result, the dependence of the temperature T on the stress σ is fixed by equality:

$$\sigma = \sqrt{\frac{T_e - T}{A\exp\left[-\frac{B}{(T-1)^\alpha}\right] - 1}}\,. \tag{10}$$

It is apparent that (10) cannot be solved analytically with respect to T and it requires the numerical analysis. Consequently, for further consideration we have to define the values of constants included in this equation. The analysis of expression (10) shows that at fixed relationship of parameters α, A, and B the characteristic temperature of friction surfaces T_e exists:

$$T^0 = 1 + \sqrt[\alpha]{\frac{B}{\ln A}}\,. \tag{11}$$

Higher than this value, there is a decrease of T with σ growth, while below it the increase of the temperature T occurs. In the case when parameters are such that the value T^0 is negative or complex, at the increase of stress σ the lubricant temperature T grows monotonously, so that always $T > T_e$. Undoubtedly, the presence of characteristic value T^0 presents a great interest for further analysis.

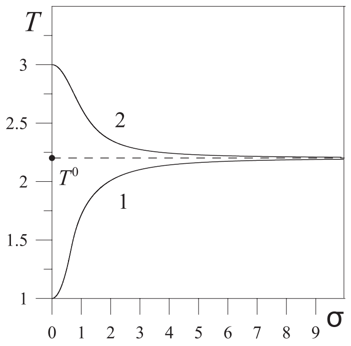

Figure 1. The dependence of the lubricant temperature on the shear stress (10) at α=A=2, $B = 1$. The curves 1 and 2 correspond to the temperatures $T_e = 1$ and 3, respectively.

Let us assume $\alpha = 2$, since at the odd values α the T vs σ dependence is two-valued at small σ which complicates the further analysis. Then, according to (11), for T^0 existing, the condition $A > 1$ has to fulfill. At $A = 2$ and $B = 1$ the formula (10) assumes the form shown in figure 1, where the curve 1 corresponds to the value $T_e < T^0$, and the curve 2 — $T_e > T^0$. The dashed line in figure $T_e = T^0$ is the asymptote for the indicated dependencies, because with the increase of σ the temperature of the system T goes to the value T^0.

The dependence of the strain on the stress $\varepsilon(\sigma)$ is fixed by equality

$$\varepsilon(\sigma) = \sigma + \frac{T_e - T(\sigma)}{\sigma}\,, \tag{12}$$

where $T(\sigma)$ is defined by relationship (10). It means that function $\varepsilon(\sigma)$ (12) allows us to carry out the numerical analysis only. At small values of sheared surface temperature $T_e < T^0$, the Hooke law is observed $\varepsilon \sim \sigma$ in the region $\sigma \ll 1$. At the σ increase, the strain decreases to zero[1], and further we again have the linearly increasing section. At $T_e \geqslant T^0$ the ε vs σ dependence has a monotonously increasing character accepting the linear form at small and large values of stress.

After substitution of (12) in (2) the Landau–Khalatnikov equation is obtained

$$\tau_\sigma \dot{\sigma} = -\partial V / \partial \sigma, \tag{13}$$

where the synergetic potential has the form

$$V = \frac{1}{2}(1-g)\sigma^2 + g \int\limits_0^\sigma \frac{T(\sigma) - T_e}{\sigma} \mathrm{d}\sigma, \tag{14}$$

in which $T(\sigma)$ dependence is defined by relationship (10). In the steady state the condition $\dot{\sigma} = 0$ is fulfilled and the potential (14) takes the minimum value. For the temperatures T_e less than those determined by equality

$$T_{c\,0} = 1 + \frac{1}{\sqrt{\ln 2g}}\,, \tag{15}$$

this minimum corresponds to the stress $\sigma = 0$. In that case the melting does not take place and the solid-like state of lubricant is realized. Otherwise, at $T_e > T_{c\,0}$ the stationary value of the stress differs from zero:

$$\sigma_0 = \left(\frac{g(T_e - T_0)}{1 - g} \right)^{1/2}, \qquad 0.5 < g < 1, \tag{16}$$

[1]The decreasing section of the ε vs σ dependence has no physical meaning.

and increases with T_e in accordance with the root law. This causes the lubricant melting and its transition into the liquid-like state. The corresponding strain is equal to

$$\varepsilon_0 = \left(\frac{T_e - T_0}{g(1-g)}\right)^{1/2}. \tag{17}$$

According to equations (10), (16) the stationary temperature T_0 coincides with the critical value T_{c0}, but differs from the characteristic value of temperature T^0 (11), except the particular case $g = 1$. Since T_{c0} is the minimum value of temperature at which the melting begins, the above means that the negative feedback of the stress σ and the strain ε on the temperature T (see the third term on the right-hand side of (4)) reduces the film temperature so much that only in this limit it ensures the self-organization process. According to (15), if the constant $g \leqslant 0.5$, the solid-like–liquid-like transition does not take place, because the temperature T_{c0} does not exist. The dependencies of the stationary shear stress on the temperature of friction surfaces are shown in figure 2 for different g parameters. They meet the second-order phase transition representing the melting of amorphous lubricant. It is worth noting that in work [4] we have obtained the value $T_{c0} = 1 + g^{-1}$ substantially differing from (15).

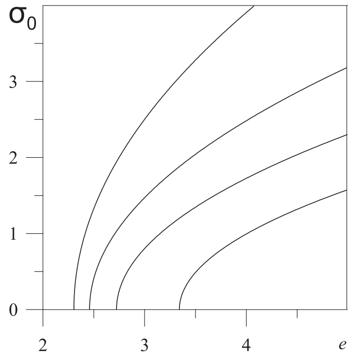

Figure 2. The dependence of the stationary values of the shear stress σ_0 on the temperature T_e (16). The curves, located from right to left, correspond to the values $g = 0.6$, 0.7, 0.8, 0.9.

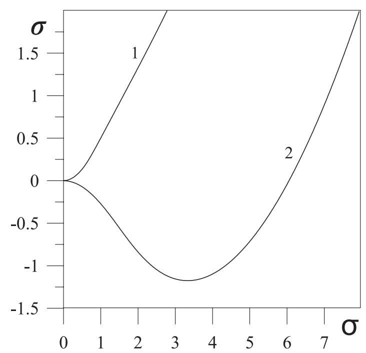

Figure 3. The dependence of the synergetic potential V (14) on the shear stress σ at $g = 0.8$, $\alpha = A = 2, B = 1$. The curves 1 and 2 correspond to the values $T_e = 1$ and 5, respectively.

The dependence of the synergetic potential V (14) on the stress σ is shown in figure 3. The curve 1 meets the temperature values $T_e < T_{c0}$ and corresponds to the solid-like state of lubricant, because the minimum of potential is realized at $\sigma = 0$. This is apparent from figure 2, in which at $T_e < T_{c0}$ the stationary stress takes only zero value. The curve 2 is built at $T_e > T_{c0}$ and corresponds to the liquid-like state of lubricant, since here the non-zero minimum of potential is realized.

At consideration of general case, without fixing the concrete values α, A, and B, the function $T(\sigma)$ is defined by dependence (10) in all previous expressions, and the critical temperature $T_{c0} = T_0$ assumes the form:

$$T_{c0} = 1 + \sqrt[\alpha]{\frac{B}{\ln gA}}. \tag{18}$$

It is apparent that at the even values of α for the temperature T_{c0} existing, the condition $gA \equiv \tau_\varepsilon G/\eta_0' > 1$ has to fulfill. In that case the value T_{c0} is always positive and it decreases with the increase of the value of the product gA. Consequently, the reduction of friction should be expected in the systems with the large gA value. However, since the radical brings a positive contribution to the expression (18), in this case the critical temperature does not decrease below the value $T_{c0} = 1$.

At odd α the temperature T_{c0} exists at all positive values of g and A. Besides, the radical can provide a negative contribution to the expression (18) leading to the values of temperature

$T_{c\,0} < 0$. The positive temperature $T_{c\,0}$ is realized at $gA > 1$ (when the radical gives a positive contribution), or in the case $gA < 1$ at the fulfillment of condition

$$|\ln gA| > B. \tag{19}$$

In the limit $B = -\ln gA$ the lubricant melts even at zero temperature of friction surfaces T_e and its liquid-like state is always realized.

4. Power dependence

In the previous section the temperature dependence of viscosity $\eta(T)$, fixed by Vogel-Fulcher relationship (7), is considered. Let us use the power dependence for the approximation of $\eta(T)$ (6). In that case in the initial system (2)–(4) the second equation assumes the form

$$\tau_\varepsilon \dot{\varepsilon} = -\varepsilon + (T-1)^\gamma \sigma. \tag{20}$$

Then, within the framework of adiabatic approximation (9) we obtain the $T(\sigma)$ dependence

$$\sigma = \sqrt{\frac{T_e - T}{(T-1)^\gamma - 1}}\,, \tag{21}$$

which at arbitrary γ qualitatively repeats the equation (10) represented in figure 1. There is only one difference in this case – the characteristic value of temperature T^0 is fixed by equality $T^0 = 2$. The $\varepsilon(\sigma)$ dependence is found from condition (12), where $T(\sigma)$ is given by expression (21), and it has the form described in section 3.

Similarly, we obtain the Landau-Khalatnikov equation (13) with synergetic potential (14), in which $T(\sigma)$ is defined by the relationship (21). In this case the critical temperature

$$T_{c\,0} = 1 + g^{-1/\gamma} \tag{22}$$

coincides with its stationary value T_0, as at use of Vogel-Fulcher formula. Note that in (22) the parameter $g \equiv G/G_0$ may be arbitrary. The stationary values of the stress and the strain are given by relationships (16), (17). Thus, at power dependence $\eta(T)$ the similar behaviour of the system is observed differing only in the values of critical $T_{c\,0}$, stationary T_0, and characteristic T^0 temperatures.

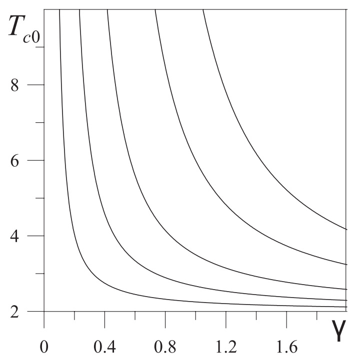

Figure 4. The dependence of the critical temperature $T_{c\,0}$ on the parameter γ (22). The curves, located from right to left, correspond to the values $g = 0.1, 0.2, 0.4, 0.6, 0.8$.

The dependencies of the critical temperature $T_{c\,0}$ on the parameter γ are presented in figure 4 for the different values $g < 1$. It is seen that $T_{c\,0}$ decreases with the growth of γ and g. At large γ values the temperature $T_{c\,0}$ asymptotically approaches the minimum possible value $T_{c\,0} = T^0 = 2$. Consequently, since the lubricant melting occurs above the temperature $T_{c\,0}$, the reduction of friction should be expected in the systems with the large values of $g < 1$ and $\gamma > 0$. At small γ the critical temperature $T_{c\,0}$ diverges and the lubricant represents a solid-like structure at arbitrary real temperatures T_e of friction surfaces.

5. The effect of deformational defect of modulus

Actually, the shear modulus, introduced (in terms of the relaxation time τ_σ) in the equation (2), depends on the stress value. This leads to the transition of the elastic deformation mode to the plastic one. It takes place at characteristic value of the stress σ_p, which does not exceed the value σ_s (otherwise the plastic mode is not manifested). In considering the deformational defect of the modulus we shall use $\tau_\sigma(\sigma)$ dependence proposed in [4] instead of τ_σ. Thus, equation (2) takes the form:

$$\tau_p \dot{\sigma} = -\sigma \left(1 + \frac{\theta^{-1} - 1}{1 + \sigma/\alpha}\right) + g_\Theta \varepsilon, \tag{23}$$

where the relaxation time for the plastic mode $\tau_p = \eta_\sigma/\Theta$ is introduced ($\eta_\sigma \equiv \tau_\sigma G$ is the effective viscosity, Θ is the hardening factor), $\theta = \Theta/G < 1$ is the parameter describing the ratio of tilts of the plastic and the Hookean sections of the deformation curve, $g_\Theta = G^2/\Theta G_0$ and $\alpha = \sigma_p/\sigma_s$ are the constants. Then, for Vogel-Fulcher case, within the framework of approximation (9) the Lorentz system (23), (8), and (4) is reduced to the Landau-Khalatnikov equation (13) with τ_p instead of τ_σ. However, in synergetic potential (14) the coefficient $g = G/G_0$ is replaced by g_Θ, which is formally assumed to be independent of σ, and the odd term appears proportional to $\theta^{-1} - 1$:

$$V = \frac{1}{2}(1 - g_\Theta)\sigma^2 + g_\Theta \int\limits_0^\sigma \frac{T(\sigma) - T_e}{\sigma} \mathrm{d}\sigma + \alpha^2(\theta^{-1} - 1)\left[\frac{\sigma}{\alpha} - \ln(\alpha + \sigma)\right]. \tag{24}$$

The consideration of the case $\dot{\sigma} = 0$ leads to the equation for the stationary values of the shear stress σ_0:

$$\sigma_0^3(g - 1) + \sigma_0^2(g - \theta^{-1})\alpha + \sigma_0(T_e - T_0)g + g\alpha(T_e - T_0) = 0. \tag{25}$$

The connection of the stationary stress and temperature $T_0(\sigma_0)$ is fixed by equation (10), where $T = T_0$. Equation (25) has the third power with respect to σ_0. However, it cannot be analysed analytically, because here the quantity T_0 is defined by more complex relationship obtained at substituting the expression (10) into (25).

In the case of power dependence $\eta(T)$ we arrive at potential (24), in which $T(\sigma)$ function is defined by (21). The consideration of the stationary situation gives an equation for the shear stress σ_0 (25), where the expression for the stationary temperature follows from (21) at $\sigma \equiv \sigma_0$. In this case we cannot obtain the analytical expression for T_{c0} either. Fixing the set of specific parameters, it is possible to carry out only the numerical analysis of the obtained equations for both dependencies.

6. Conclusion

The above consideration shows that the increase of temperature of the rubbing surfaces T_e can be accompanied by self-organization of the system leading to the sliding friction mode. If the Vogel-Fulcher temperature dependence of viscosity is valid, it is possible to select the three classes of the systems on the lubricant state depending on temperatures of the sheared surfaces:

- the lubricant has the solid-like structure at arbitrary temperature of friction surfaces;
- there is the critical value of temperature, above which the lubricant melting occurs and it passes from solid-like into liquid-like state;
- the lubricant is liquid-like even at zero temperature of friction surfaces.

In the case when the dependence of viscosity on the temperature is fixed by power expression, the minimum possible value of the rubbing surfaces temperature, at which the lubricant melts, is limited by the value $T_{c0} = 2$ at $g \equiv G/G_0 \to 1$ or $\gamma \to \infty$. Obviously, the first type of lubricating materials is most preferable in the concrete devices and mechanisms, since the conditions for friction reduction are easier to provide. Taking into account the deformational defect of the modulus we have obtained the equations describing the crystalline lubricant melting.

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Вплив температурної залежності в'язкості на плавлення ультратонкої плівки мастила

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Досліджується плавлення ультратонкої плівки мастила в процесі тертя між атомарно-плоскими поверхнями при температурних залежностях в'язкості, які описуються співвідношенням Фогеля-Фулчера і степепеневим виразом, що спостерігаються експериментально. Показано, що в обох випадках існує критична температура, при перевищенні якої відбувається плавлення плівки мастила, і як наслідок встановлюється рідинний режим тертя. Визначені значення характерних параметрів мастила, необхідні для зменшення тертя. У системах, де виконується залежність Фогеля-Фулчера, можливо підібрати такі параметри, при яких плавлення мастила відбувається навіть при нульовій температурі поверхонь тертя. Для опису процесу плавлення мастила за механізмом переходу першого роду проведено врахування деформаційного дефекту модуля зсуву.

Ключові слова: *в'язкопружне середовище, граничне тертя, зсувні напруження та деформація*

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