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NON-EQUILIBRIUM STATIONARY MODES OF BOUNDARY FRICTION

***I.A. Lyashenko*[1], *L.S. Metlov*[2], *A.V. Khomenko*[1], *S.N. Chepulskyi*[1]**

[1] Sumy State University,
2, Rimsky-Korsakov Str., 40007 Sumy, Ukraine
E-mails: nabla04@ukr.net, khom@mss.sumdu.edu.ua, apichsn@gmail.com

[2] Donetsk Institute for Physics and Engineering,
named after A.A. Galkin of the NASU,
72, R.-Luxemburg Str., 83114 Donetsk, Ukraine
E-mail: lsmet@fti.dn.ua

A non-equilibrium thermodynamic model describing the behavior of an ultrathin lubricating material film confined between two atomically-smooth solid surfaces is proposed. As the order parameter the excess volume is chosen whose value increases with melting and subsequent fluidization of lubricant. It is shown that the external pressure, applied perpendicularly to the friction surfaces, influences crucially the lubricant behavior. The total system of kinetic equations is written down in which the relative shear velocity of rubbing surfaces, their temperature, and external normal pressure are the control parameters. Phase diagram with domains of different friction regimes is built.

Keywords: *BOUNDARY FRICTION, NON-EQUILIBRIUM PROCESSES, INTERNAL ENERGY, STICK-SLIP MODE, INVARIANTS OF STRAIN TENSOR, SHEAR MELTING.*

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1. INTRODUCTION

The boundary friction mode occurring at the lubricant thickness less than 10 atomic layers [1] has been actively studied during last years with the swift development of the experimental investigation methods of micro- and nanosystems. In the experiments, atomically-smooth mica surfaces are often used as the friction surfaces, and spherical octamethylcyclotetrasiloxane (OMCTS) molecules and linear chain tetradecane and hexadecane molecules – as the lubricants [2, 3]. Such thin layer displays anomalous properties with respect to volume lubricants [2]. Interrupted (stick-slip) motion mode [2-5] is one of the peculiarities inherent in a dry friction [4]. The mentioned mode arises due to the periodic solidification of lubricant during its compression by rubbing surfaces and subsequent stepwise melting with excess of the yield strength of shear stresses. This mechanism got the name shear melting, and in such systems it appears simultaneously with the usual thermodynamic one.

During melting of an ultrathin lubricant layer its stationary states are not equilibrium thermodynamic phases, but represent kinetic modes of friction. The reason is in the fact that atomic relief of rubbing surfaces separated by the lubricant influences strongly the state symmetry of the lubricant [6]. The pressure applied to the surfaces has nontrivial effect on the establishment of the friction mode [2]. In friction, lubricant states are very much different from the solid and liquid states, in which this lubricant of larger thickness can be. Therefore, one introduces the solid-like and liquid-like phases. It is possible to judge about melting of ultrathin lubricant layers by the increase in their volume [7] and the diffusion coefficient [7-9] as well. Volume is the experimentally observed quantity, therefore to describe the system state we have introduced in [10] the parameter f, whose square has physical meaning of an excess volume arising due to the chaotization of the structure of solid during melting. With the increase in f defect density in lubricant grows, and because of this it goes to the liquid-like phase, which during friction corresponds to the kinetic mode of plastic flow or sliding of rubbing surfaces with low friction force between them.

The given paper is the continuation of works [10-13] and is devoted to the investigation of the boundary friction mode taking into account the non-equilibrium processes arising due to the energy exchange between lubricant and environment represented by rubbing surfaces, which play the role of the thermostat. Nontrivial influence of the pressure on the establishment of the friction mode, which was specified experimentally earlier [2], is also shown in the present work.

2. INTERNAL ENERGY AND STATIONARY STATES

We write the expansion for the internal energy bulk density u by the parameter f taking into account contributions from the elastic components of shear starins ε_{ij}^{e}, equilibrium s and non-equilibrium $\tilde{s}$ components of the entropy in the form of

$$u = u_0 + t_0\tilde{s} - \frac{1}{2}t_1\tilde{s}^2 + \phi_0 f^2 - \frac{1}{4}\phi_1 f^4 + \frac{1}{6}\phi_2 f^6 , \qquad (1)$$

where f^2 is the excess volume, whose zero value corresponds to the solid-like structure of lubricant and non-zero – to the liquid-like [10]. In turn,

$$u_0 = u_0^* + \frac{1}{2}\lambda\left(\varepsilon_{ii}^{e}\right)^2 + \mu\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} + \alpha s^2 , \qquad (2)$$

$$\phi_0 = \phi_0^* - \frac{1}{2}\bar{\lambda}\left(\varepsilon_{ii}^{e}\right)^2 - \bar{\mu}\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} - \alpha_{\phi}s^2 , \qquad (3)$$

$$t_0 = t_0^* + \frac{1}{2}\tilde{\lambda}\left(\varepsilon_{ii}^{e}\right)^2 + \tilde{\mu}\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} , \qquad (4)$$

where ε_{ii}^{e}, $\varepsilon_{ij}^{e}\varepsilon_{ji}^{e}$ are the first two strain tensor invariants.

Elastic stresses are expressed through the internal energy (1)

$$\frac{\partial u}{\partial \varepsilon_{ij}^{e}} \equiv \sigma_{ij}^{e} = \lambda\varepsilon_{ii}^{e}\delta_{ij} + 2\mu\varepsilon_{ij}^{e} - \left(\bar{\lambda}\varepsilon_{ii}^{e}\delta_{ij} + 2\bar{\mu}\varepsilon_{ij}^{e}\right)f^2 + \left(\tilde{\lambda}\varepsilon_{ii}^{e}\delta_{ij} + 2\tilde{\mu}\varepsilon_{ij}^{e}\right)\tilde{s} , \qquad (5)$$

where ε_{ij}^{e} is the shear component of elastic strain. The last expression can be represented in the form of the effective Hooke law [14]

$$\sigma_{ij}^{e} = 2\mu_{eff}\varepsilon_{ij}^{e} + \lambda_{eff}\varepsilon_{ii}^{e}\delta_{ij} \qquad (6)$$

with effective elastic parameters

$$\mu_{eff} = \mu - \bar{\mu} f^2 + \tilde{\mu}\tilde{s}\ , \tag{7}$$

$$\lambda_{eff} = \lambda - \bar{\lambda} f^2 + \tilde{\lambda}\tilde{s}\ , \tag{8}$$

which are decreased with the increase in the excess volume growing at the lubricant melting.

For the plain-strain lubricant state, in which $\varepsilon_{22}^{e} = 0$, the strain tensor invariants are determined as follows [11, 12, 15]:

$$\varepsilon_{ii}^{e} = \frac{n}{\lambda_{eff} + \mu_{eff}}\ , \tag{9}$$

$$\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} = \frac{1}{2}\left[\left(\frac{\tau}{\mu_{eff}}\right)^2 + \left(\varepsilon_{ii}^{e}\right)^2\right], \tag{10}$$

where n and τ are the normal and tangent components of the stresses acting on the lubricant from rubbing surfaces. Shear stress τ is found from the expression (6), which at $i \neq j$ ($\delta_{ij} = 0$) leads to the dependence

$$\tau = 2\mu_{eff}\varepsilon_{ij}^{e}\ . \tag{11}$$

At first, we consider the simplified situation, when in the expression for the internal energy (1) both entropy components are equal to zero. In Fig. 1a we present the corresponding profiles of the internal energy $u(f)$.

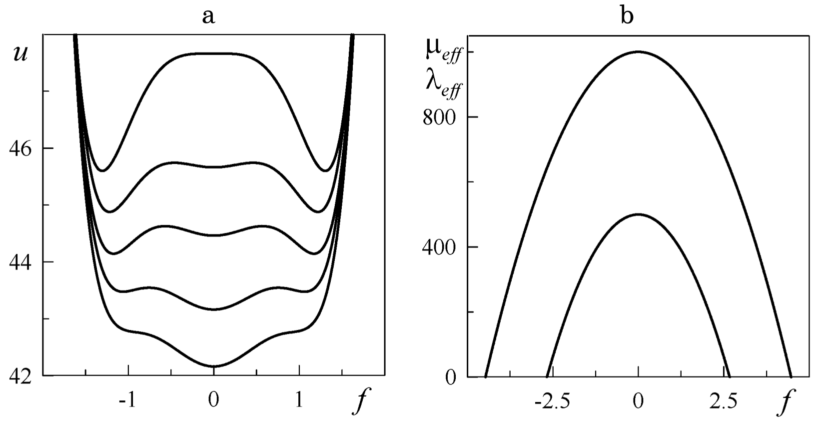

Fig. 1 *– a: dependence of the internal energy density u (J/m^3) (1) on the parameter f (dimensionless quantity) at $\phi_0^* = 2$ J/m^3, $\phi_1 = 8{,}5$ J/m^3, $\phi_2 = 5$ J/m^3, $\lambda = 5{\cdot}10^{11}$ Pa, $\bar{\lambda} = 7{\cdot}10^{10}$ Pa, $\mu = 10^{12}$ Pa, $\bar{\mu} = 5{\cdot}10^{10}$ Pa, $n = -\,7{\cdot}10^5$ Pa, $\tilde{s} = s = 0$. Curves located bottom-up correspond to the strains $\varepsilon_{ij}^{e} = 10^{-6}$; $2{\cdot}10^{-6}$; $3{\cdot}10^{-6}$; $3{,}5{\cdot}10^{-6}$; $4{,}5{\cdot}10^{-6}$ and the energy grade $u_0^* = 40$; 35; 26,3; 21; 7 J/m^3; b: dependence of the effective elastic parameters μ_{eff} (GPa) (7) (the upper curve) and λ_{eff} (GPa) (8) (the lower curve) on the parameter f*

For convenience, different values of the constant u_0^* which influences only the position of the curve with respect to the ordinate axis are chosen for each curve in the figure; u_0^* does not effect on the form of the dependence $u(f)$. At small elastic strains (lower curve), a single zero minimum of the potential $u(f)$ corresponding to the stationary state with zero value of excess volume $f^2 = 0$ is realized; in this case, lubricant is solid-like. According to Fig. 1b, maximum values of the elastic parameters correspond to zero value of f. With the increase in the strain (three middle curves in Fig. 1a) two symmetrical non-zero minimums are additionally realized on the dependence except for zero one. These minimums correspond to the liquid-like state of the lubricant and are separated from zero minimum by the potential maximums. Since non-zero minimums are separated by maximums from zero minimum, for the initial solid-like state of the lubricant in the deterministic case the system transition to the state corresponding to these minimums is impossible. At the further increase in the strain (the upper curve), dividing maximums disappear, and system by the first-order phase transformation mechanism sharply goes to the state corresponding to the minimum of energy at $f \neq 0$ (lubricant melts). If now decrease the strain, at the appearance of dividing maximums solidification of the lubricant is impossible up to their disappearance.

Stationary states of the lubricant are determined by the condition $\partial u/\partial f = 0$, which leads to the equation

$$\bar{\mu}\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} + \alpha_{\phi}s^{2} = \phi_0^{*} - \frac{1}{2}\bar{\lambda}\left(\varepsilon_{ii}^{e}\right)^{2} - \frac{1}{2}\phi_1 f^{2} + \frac{1}{2}\phi_2 f^{4} + \frac{n^{2}(\bar{\lambda} + \bar{\mu})}{\left(\lambda_{eff} + \mu_{eff}\right)^{2}}, \qquad (12)$$

which can be conveniently represented as function $\varepsilon_{ij}^{e}\varepsilon_{ji}^{e}(f, n, s, \tilde{s})$ or $s(f, n, \varepsilon_{ij}^{e}\varepsilon_{ji}^{e}, \tilde{s})$. When analyzing equation (12), we will calculate the elastic shear strain ε_{ij}^{e} in accordance with the relations (9)-(11) through the invariants

$$\varepsilon_{ij}^{e} = \frac{1}{2}\sqrt{2\varepsilon_{ij}^{e}\varepsilon_{ji}^{e} - \left(\varepsilon_{ii}^{e}\right)^{2}}\,. \qquad (13)$$

Dependences of the stationary value of excess volume $f_0^2(\varepsilon_{ij}^{e})$ at the fixed values of entropy s and $f_0^2(s)$, fixed strains ε_{ij}^{e} calculated according to the equations (12), (13), and $\tilde{s} = 0$ are shown in Fig. 2.

According to the curves 1 and 2, with the increase in the strain (entropy) from zero values excess volume at first is equal to zero (lubricant is solid-like), then its value at the strain $\varepsilon_{ij}^{e} = \varepsilon_{c0}$ (entropy $s = s_{c0}$) stepwise increases, and here lubricant melts. With the further decrease in the strain (entropy) lubricant solidifies at smaller values $\varepsilon_{ij}^{e} = \varepsilon_c^0$ ($s = s_c^0$). Thus, dependence has hysteresis behavior that corresponds to the first-order phase transitions. In accordance with Fig. 2a, with the increase in the entropy melting occurs at smaller strain value, and Fig. 2b shows the decrease in the value of s_{c0} with the growth of ε_{ij}^{e}. Curve 3 on the considered figures corresponds to the situation when during melting lubricant can not solidify any more with the decrease in the corresponding control parameter (strain in Fig. 2a or entropy in Fig. 2b) since with its decrease down to zero value potential barrier on the dependence $u(f)$ (dashed part of the dependence), which impedes lubricant to solidify again, does not disappear. There are also critical values of the system entropy and elastic strains, with excess of which at the chosen value of pressure lubricant is always liquid-like (curve 4 in both figures) that implies the realization of two symmetrical non-zero minimums on the dependence $u(f)$ separated by a single zero maximum.

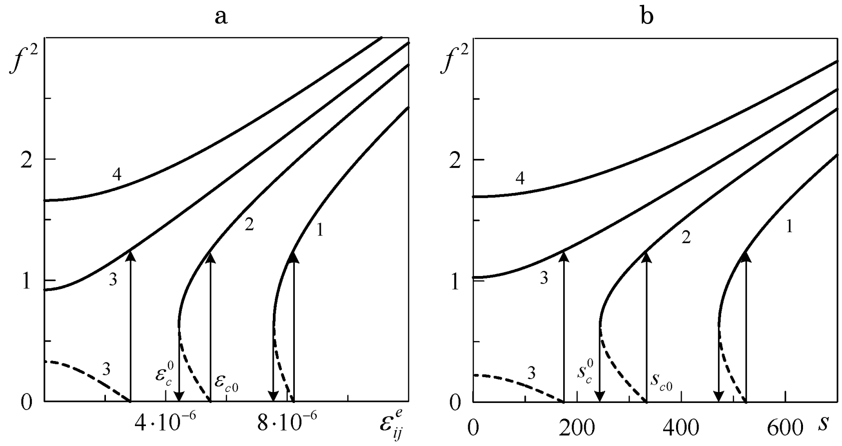

Fig. 2 *– Dependence of the stationary value of excess volume* f^2 *(dimensionless quantity) on the elastic strain* ε_{ij}^e *(dimensionless quantity) and entropy* s *(*$J \cdot K^{-1} \cdot m^{-3}$*) at the parameters of Fig. 1,* $\alpha = 0{,}95$ $K^2 \cdot m^3/J$, $\alpha_\phi = 2 \cdot 10^{-5}$ $K^2 \cdot m^3/J$, *normal stresses* $n = -1{,}5 \cdot 10^7$ *Pa, and non-equilibrium entropy* $\tilde{s} = 0$*: a – curves 1-4 correspond to the fixed values of entropy* $s = 250$*;* 500*;* 600*;* 700 *(*$J \cdot K^{-1} \cdot m^{-3}$*); b – curves 1-4 are plotted at the fixed strains* $\varepsilon = 5 \cdot 10^{-6}$*;* $7{,}6 \cdot 10^{-6}$*;* $8{,}6 \cdot 10^{-6}$*;* 10^{-5}

Fig. 3 illustrates the phase diagrams of the lubricant states in the coordinates of control parameters at $\tilde{s} = 0$ obtained due to the numerical analysis of equations (12), (13). Fig. 3a represents the dependences of the critical strains ε_{c0}, ε_c^0 on the system entropy s. Curves in this figure can be also interpreted as the dependences of s_{c0}, s_c^0 on the strain level ε_{ij}^e. Lubricant is liquid-like above the curve $\varepsilon_{c0}(s_{c0})$, and the sliding friction (SF) mode is realized. In this region potential has the form shown by the upper curve in Fig. 1a. Below the curve $\varepsilon_c^0(s_c^0)$ lubricant is solid-like that corresponds to the form of the internal energy $u(f)$ shown by the lower curve in Fig. 1a. In the region between the curves in Fig. 3 lubricant state depends on the initial conditions. Potential in the specified region is represented by three middle dependences in Fig. 1a. Regions of the phase diagram at the decreased value of normal stresses are shown by the dashed lines. One can conclude that with the increase in the load on rubbing surfaces the region of dry friction is expanded and the hysteresis region, which separates the mentioned region from the region of liquid-like state, is narrowed. The external load described in our model by normal stresses n often crucially influences the tribological nanosized system. For example, it is shown in the experimental work [2] that the load growth for some lubricant types leads to the increase in the critical melting velocity, and for other lubricants this velocity decreases. Also, pressure variations lead to the change in the phase transition frequency, etc. In Fig. 3b, c we present the phase diagrams in the pressure-strain and pressure-entropy coordinates, respectively, to ascertain the influence of external stresses on the lubricant state.

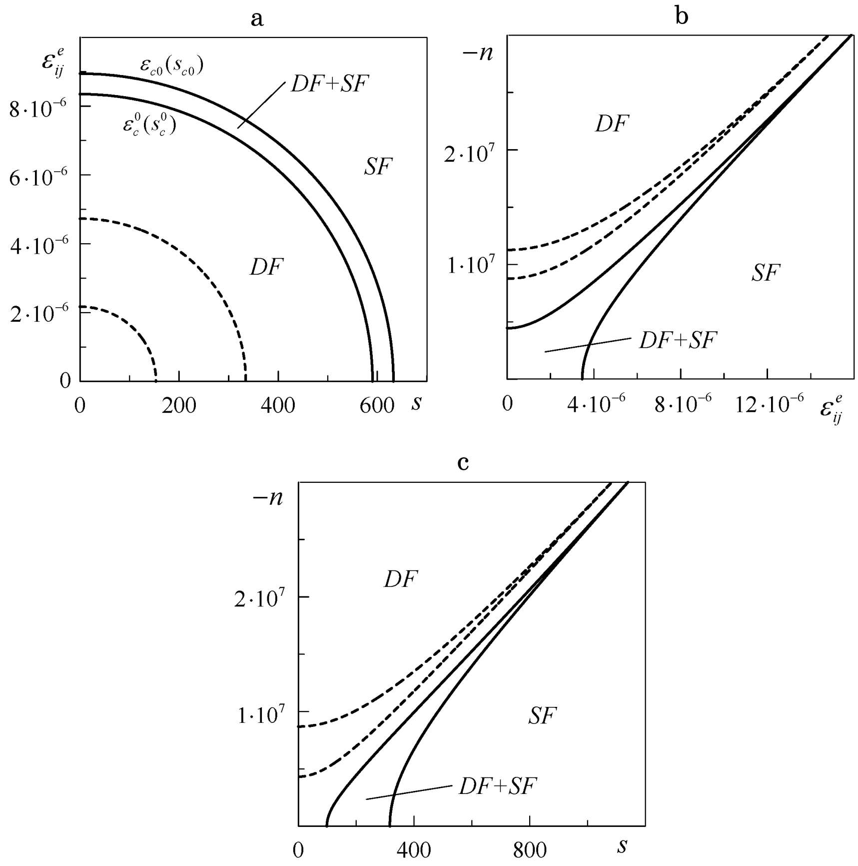

Fig. 3 *– Phase diagrams with the sliding friction (SF) and dry friction (DF) regions at the parameters of Fig. 1 and Fig. 2. a : solid lines correspond to the stress* $n = -1{,}5{\cdot}10^7$ *Pa, for dashed lines* $n = -3{\cdot}10^6$ *Pa; b – solid lines correspond to the entropy* $s = 200$ $J{\cdot}K^{-1}{\cdot}m^{-3}$*, for dashed lines* $s = 450$ $J{\cdot}K^{-1}{\cdot}m^{-3}$*; c – solid lines correspond to the strain* $\varepsilon_{ij}^e = 0$*, for dashed lines* $\varepsilon_{ij}^e = 5{\cdot}10^{-6}$

As follows from these diagrams, with the increase in the load on rubbing surfaces transition of the lubricant from the solid-like state (DF region) to the liquid-like (SF region) occurs omitting the hysteresis region (DF + SF). In other words, at large values of the external pressure the first-order phase transition is not realized, instead of it continuous lubricant melting by the second-order phase transformation mechanism takes place. Within such model [12] it was earlier shown that increase in the pressure leads to the lubricant solidification and increase in the total friction force value, and depending on the value of n both the first- and the second-order phase transitions are implemented. The model [12] describes only the first-order phase transition at small values of the external pressure.

3. KINETIC EQUATIONS

Usually in the experiments one fixes not the entropy but its change calculated using indirect measurements. Therefore, it is more convenient to fix the lubricant temperature, which can be measured directly. The temperature is determined through the internal energy (1)

$$\frac{\partial u}{\partial s} \equiv T = 2s(\alpha - \alpha_\phi f^2)\,, \tag{14}$$

whence the condition of temperature positivity gives the maximum possible value for the excess volume parameter $f_{\max} = \sqrt{\alpha/\alpha_\phi}$. Similarly we will determine the temperature of non-equilibrium subsystem

$$\frac{\partial u}{\partial \tilde{s}} \equiv \tilde{T} = t_0 - t_1\tilde{s}\,. \tag{15}$$

We write the evolution equations for non-equilibrium parameters f and $\tilde{s}$ in the Landau-Khalatnikov form

$$\tau_f \dot{f} = -\frac{\partial u}{\partial f}\,, \quad \tau_s \dot{\tilde{s}} = \frac{\partial u}{\partial \tilde{s}}\,, \tag{16}$$

where constants τ_f, τ_s are inversely proportional to the kinetic coefficients. According to equations (16) internal energy by the coordinate f tends to its minimum value, and by the coordinate $\tilde{s}$ – to the maximum that corresponds to non-equilibrium processes [16, 17].

Evolution equations (16) take the following explicit form:

$$\tau_f \frac{\partial f}{\partial t} = -2\phi_0 f + \phi_1 f^3 - \phi_2 f^5 - \frac{2n^2(\bar{\lambda} + \bar{\mu})f}{(\lambda_{eff} + \mu_{eff})^2}\,, \tag{17}$$

$$\tau_s \frac{\partial \tilde{s}}{\partial t} = t_0 - t_1\tilde{s}\,, \tag{18}$$

where presence of the last term in (17) is conditioned by the dependence of the invariants (9), (10) on the value of f. In equation (18) term with the sign "+" describes the non-equilibrium entropy increment due to external energy sources (work) and with the sign "–" – its escape to equilibrium subsystem.

Since we consider the non-equilibrium open system, to describe the heat exchange processes between the lubricant and environment it is necessary to introduce the temperature of rubbing surfaces T_e. In the case of non-uniform heating of the environment the heat conduction equation is usual continuity equation [14]

$$T\frac{\partial s}{\partial t} = \kappa \nabla^2 T\,, \tag{19}$$

where the heat conductivity coefficient κ is assumed to be constant. To simplify the problem we will consider the lubricant to be uniform over the plane ($\nabla_x^2 = \nabla_y^2 = 0$). In view of thinness of the lubricant for the normal component

∇_z^2 with satisfactory accuracy one can use the approximation $\nabla_z^2 T \approx (T_e - T) / h^2$, where h is the lubricant thickness or the distance between rubbing surfaces. Taking into account this equation, (19) will be written in the simpler form

$$\frac{\partial s}{\partial t} = \frac{\kappa}{h^2}\left(\frac{T_e}{T} - 1\right), \tag{20}$$

where h^2/κ plays the role of the relaxation time, during which temperature equalization in the lubricant thickness due to the heat conduction occurs. However, (20) does not yet take into account the interaction of equilibrium and non-equilibrium subsystems. Non-equilibrium entropy loss is taken into consideration by negative terms in the evolution equation (18). This means that these terms should take into account the equilibrium entropy increment. With account for this circumstance we will obtain the final equation for the equilibrium entropy

$$\tau_s \frac{\partial s}{\partial t} = t_1 \tilde{s} + \tau_s \frac{\kappa}{h^2}\left(\frac{T_e}{T} - 1\right). \tag{21}$$

In this case, the lubricant temperature T (14) in the stationary state $\partial s/\partial t = 0$ is determined not only by the temperature of rubbing surfaces T_e, which plays the thermostat (environment) role as it was in (20), but also by the non-equilibrium entropy value $\tilde{s}_0$.

In [10-13] we have obtained the kinetic equation for the strain ε_{ij}^e in the form of

$$\tau_\varepsilon \dot{\varepsilon}_{ij}^e = -\varepsilon_{ij}^e + \frac{V_{ij}\tau_\varepsilon}{h}, \tag{22}$$

where τ_ε is the Maxwell relaxation time of internal stresses, and V_{ij} is the relative shear velocity of rubbing surfaces.

To study the system kinetics it is necessary to simultaneously numerically solve the set of kinetic equations (17), (18), (21), (22) determining invariants according to (9), (10), shear stresses τ from (11), elastic constants from (7), (8), and current lubricant temperature T from (14).

According to the equations given in this Section, the control parameters which can be specified arbitrary are the following: temperature of rubbing surfaces T_e, load on the upper rubbing surface n, and velocity of rubbing surfaces V_{ij}, whose value determines the strain level ε_{ij}^e. Stationary values of the lubricant temperature T, entropy s, non-equilibrium entropy $\tilde{s}$, as well as the excess volume f^2 are established in time as a result of system evolution in accordance with the chosen control parameters [13]. In general case, this complicates the consideration carried out in the previous Section where the mentioned parameters are assumed to be independent and chosen arbitrary. Moreover, accounting of non-equilibrium entropy assumes larger number of stationary states that it is shown in Fig. 3. For example, the stationary condition $\partial\tilde{s}/\partial t$ in (18) leads to the equation for stationary states

$$\left[\mu + \lambda - f^2(\bar{\mu} + \bar{\lambda}) + \tilde{s}(\tilde{\mu} + \tilde{\lambda})\right]^2\left(t_1\tilde{s} - t_0^* - 2\tilde{\mu}(\varepsilon_{ij}^e)^2\right) - \frac{1}{2}(\tilde{\lambda} + \tilde{\mu})n^2 = 0, \tag{23}$$

where ε_{ij}^{e} is the elastic strain. Equation (23) with respect to $\tilde{s}$ is a cubic one and has non-zero coefficients at all degrees of $\tilde{s}$, it also contains a free term. In general case, it has three roots, which represent the stationary values of $\tilde{s}_0$. At that the picture of stationary states shown in Fig. 3 where $\tilde{s} = 0$ is considerably complicated.

4. CONCLUSIONS

In the present work, a general theory which adequately describes phenomena occurring during boundary friction is developed. Thermodynamic (with the increase in the system entropy) and shear (with the increase in the strain) melting is considered. It is shown that elastic moduli decrease with the excess volume growth that leads to the lubricant fluidization in dynamic systems. Stationary states are considered, however, closed system of kinetic equations which can be used for the study of the process dynamics and description of existing experimental data is also written in the work. When constructing the model, the heat conduction processes are taken into account that allows to describe the lubricant solidification due to heat transfer to rubbing surfaces (environment). Two subsystems are marked out, namely, the non-equilibrium subsystem which obtains heat during the work on a system (during surface shear) and the equilibrium one, in which this heat partially passes due to the internal processes. Thus, an open system with non-equilibrium type connected with two-way energy exchange between system and environment is considered.

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