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Analytical calculation of self-oscillatory dynamics of neck propagation in polymers

© *A.A. Chevrychkina*[1], *A.L. Korzhenevskii*[1,¶]

[1] Federal State Budgetary Institution of Science Institute of Machine Science, Russian Academy of Sciences, St. Petersburg, Russia

[¶] E-mail:alekorzh@mail.ru

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A method for analytical calculation of parameters of self-oscillations of stress, temperature, and neck advance velocity in a polymer film stretched with a given constant velocity is proposed. It is shown that in the most common case, when high sample compliance is required to generate self-oscillations, the dynamics of self-oscillations can be described by an ordinary nonlinear differential equation of the generalized Lienard equation type. An exact relationship is revealed between the non-stationary dynamics of the polymer film neck front and the explosive crystallization front propagating in the self-oscillation mode.

Keywords: self-oscillations of a polymer film, thermal model, derivation of an ordinary nonlinear differential equation.

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1. Introduction

The existence of the mode of self-oscillations of polymer neck front (NF) in the process of cold drawing was detected in independent papers [1–3]. The first theoretical model [4] explained this dynamic effect by the presence of the positive feedback causing thermomechanical instability.

The multiple subsequent experiments thoroughly studied the terms necessary for the occurrence of the effect, depending on the specified values of deformation rate, temperature, dimensions and geometry of samples, and also compliance of the experimental setup [5–31]. It was found that the effect is observed in some polymers having a qualitatively different initial internal structure. In most materials it is accompanied with temperature increase by several dozens of degrees in the narrow zone of NF. The contributory factor is the occurrence of the pores, the concentration of which may be significant. Inside the self-oscillation area the bifurcation points were found, which, when passed, changed the periods of self-oscillations by a multiple number of times.

Compared to the broad set of the accumulated experimental data, the progress in the theoretical studies was of much more limited nature. Further development of the knowingly strongly simplified model [4] was carried out in papers [12,14,25,26]. At the same time they maintained the main assumption of the dominant effect of thermal positive feedback for occurrence of the self-oscillatory mode known as a „thermal" model (TM).

Besides, the strong nonlinearity of the currently long-present version of TM resulted in the fact that it was only possible to analytically calculate the stationary motion mode of NF within its framework [12,25], whereas the calculation of the boundary of its instability region was implemented inconsistently, which led to the wrong definition of its position in [22] (see formula (4) in [22]).

For the quantitative calculations of the non-linear stage of front advancement, and the time dependences of the stress and temperature thereon, only the numerical methods had to be used [19,20]. The results of the latter are obviously useful when compared to the data obtained for a specific polymer with the fixed parameters of the experiment realization, but they will not say much about the universal applicability or inapplicability of the TM in general in virtue of multiple initial parameters included therein.

As for the alternative proposal on the leading role of the formation of shear bands and pores in their vicinity in the occurrence of the self-oscillatory mode, no adequate mathematical model has yet been offered [31]. In this article we will not consider the self-oscillations provided for by non-thermal mechanisms.

From the above it is clear that the deep understanding of the combination of available experimental results and the mechanisms they are based upon, is delayed by the absence of the analytical calculations of the non-stationary dynamics of the neck advancement, with which the quantitative comparison of experimental data could be done. It is evident that such calculations will also be useful for the interpretation of the results produced purely by the computer-aided method.

In this paper we will show that in the most typical experimental situation, when it is necessary to artificially increase the system compliance for excitation of self-oscillations, the TM dynamics may be precisely described by the ordinary differential equation for the non-linear oscillator that has negative friction. The coefficients of this oscillator containing the alternating effective mass and friction force may be fully expressed via the parameters known experimentally.

2. Formulation of the model

„The canonical" TM of the polymer film is described by the system made of 3 equations [12,13]:

$$\frac{\partial T}{\partial t} = \eta \frac{\partial^2 T}{\partial x^2} - \Gamma(T - T_0) + \frac{\lambda - 1}{\rho c} \sigma(V, T) V(t) \delta\big(x - X(t)\big), \tag{1}$$

$$\dot{\sigma} = \frac{V_p - (\lambda - 1)V}{D}, \tag{2}$$

$$V = V_0 \exp\left(\frac{a\sigma - U}{RT}\right). \tag{3}$$

In the thermal conductivity equation (1) $T(x,t)$ — local temperature of the film, T_0 — ambient air temperature, σ — elastic stress, $V(t) \equiv \dot{X}(t)$ — NF velocity, $X(t)$ — coordinate of its current position, η — thermal diffusivity coefficient, Γ — coefficient of the film heat transfer into the environment, ρ and c — density and heat capacity, $\delta(x)$ — Dirac's delta function. Modelling of the source in the equation (1) by this function reflects the experimental fact that the width of the transition zone of heat release is much smaller than the remaining lengths of the task.

Equation (2) expresses the condition that the sum of the deformation rates of the elastic (non-oriented) part of the film and its plastically deformed (oriented) part is equal to the specified constant tensile speed V_p. In (2) the point above the stress $\sigma(t)$ indicates the differentiation by time t, coefficient D — elastic compliance of the machine + sample system (for„the rigid" machine $D = L/E$, where L — film length, E — modulus of elasticity), coefficient λ describes the extent of the polymer drawing in the neck. Its value, as well as the value of the free volume a in equation (3), increases noticeably with the temperature after the film heating beyond the glass transition temperature, but to simplify the formulas, let us assume that λ and a are the constant parameters, and we will cover the influence of temperature dependence briefly in the end of the article.

The phenomenological equation (3) describes the rate of the plastic deformation selected as the Arrhenius formula.

3. Transition to dimensionless variables and parameters

To simplify the appearance of the formulas in the subsequent calculations, and also for greater clarity of the below common results and to detect the small parameters, it is feasible to use some dimensionless parameters and functions.

Let us introduce the dimensionless coordinate $\tilde{x}$, time $\tilde{t}$, stress and temperature:

$$\tilde{x} = \frac{x}{\frac{4\eta}{V_s}}, \quad \tilde{t} = \frac{t}{\frac{4\eta}{V_s^2}}, \quad \widetilde{\sigma} = \frac{\sigma}{\sigma_s}, \quad \widetilde{T} = \frac{T}{T_s}. \tag{4}$$

Then the dimensionless speed is $\widetilde{V} = \frac{V}{V_s}$, where $V_s = \frac{V_p}{\lambda - 1}$ — NF speed in the stationary mode.

Let us also introduce the dimensionless parameters:

$$\alpha = C \frac{\sigma_s}{T_s} \ln\left(\frac{V_0}{V_s}\right)\sqrt{\beta}, \ \beta = \frac{V_s^2}{V_s^2 + 4\eta\Gamma}, \ \gamma = a\frac{\sigma_s}{RT_s},$$
$$\delta = \frac{2\eta}{V_s \sigma_s} \frac{(\lambda - 1)}{D}, \quad \epsilon = \ln\left(\frac{V_0}{V_s}\right). \tag{5}$$

The system (1) in the dimensionless variables and parameters (4), (5) has the following appearance:

$$\begin{cases} \dot{\widetilde{T}} = \dfrac{1}{4} \dfrac{\partial^2 \widetilde{T}}{\partial \tilde{x}^2} - \dfrac{1-\beta}{\beta}\left(\widetilde{T} - \left(1 - \dfrac{\alpha}{\epsilon}\right)\right) \\ \qquad + \dfrac{\alpha}{\epsilon\sqrt{\beta}} \widetilde{\sigma}\widetilde{V}\delta(\tilde{x} - \widetilde{X}), \\ \dot{\widetilde{\sigma}} = 2\delta(1 - \widetilde{V}), \\ \ln(\widetilde{V}) - \epsilon = \dfrac{\gamma\widetilde{\sigma} - (\gamma + \epsilon)}{\widetilde{T}}. \end{cases} \tag{6}$$

4. Approach to the effective solution to the non-stationary equation of thermal conductivity in polymers

The main difficulty that until now prevented the generation of analytical results for calculation of the non-stationary dynamics of system (6), was due to the fact that the trajectory $\widetilde{X}(\tilde{t})$ for NF advancement is not known in beforehand, therefore it was necessary to search for the solution to the thermal conductivity equation at the arbitrary dynamics of the source in (6_1). We will demonstrate how this difficulty may be overcome, and how the analytical solution of the system (6) may be obtained in that practically important case, when the parameter δ must be decreased to excite the self-oscillations. As it follows from (5), the small parameter δ complies with the high values of compliance and/or tensile speed, when the self-oscillations are usually observed.

Let us write the solution to the thermal conductivity equation (6_1) in the form of the Green's function convolution with the heat source. Temperature at NF with coordinate $\widetilde{X}(\tilde{t})$ is equal to:

$$\widetilde{T} - \left(1 - \frac{\alpha}{\epsilon}\right) = \frac{\alpha}{\epsilon\sqrt{\beta}} \int_{-\infty}^{\tilde{t}} d\tilde{t}'\, G\big(\widetilde{X}(\tilde{t}) - \widetilde{X}(\tilde{t}'), \tilde{t} - \tilde{t}'\big)$$
$$\times\, \widetilde{\sigma}\Big(\widetilde{V}(\tilde{t}'), \widetilde{T}\big(\widetilde{X}(\tilde{t}')\big)\Big)\widetilde{V}(\tilde{t}'), \tag{7}$$

where the Green's function is determined as:

$$G\big(\widetilde{X}(\tilde{t}) - \widetilde{X}(\tilde{t}'), \tilde{t} - \tilde{t}'\big) = \frac{1}{\sqrt{\pi(\tilde{t} - \tilde{t}')}}$$
$$\times \exp\left\{-\frac{1-\beta}{\beta}(\tilde{t} - \tilde{t}') - \frac{[\widetilde{X}(\tilde{t}) - \widetilde{X}(\tilde{t}')]^2}{(\tilde{t} - \tilde{t}')}\right\}. \tag{8}$$

In the right part of the formula (7) we neglected the additional contribution related to the temperature variation due to the difference in the tensile speed of the neck material and the NF spread. A simple estimate shows that the relative value of this contribution is equal to $\frac{1-\beta}{4\beta(\lambda-1)}$ and is only several percents at all speeds, except for the lowest ones. But in the last case the film heating becomes insignificant, and it no longer makes sense to use the activation TM.

We make it clear that the formula (7) as such is not a real solution to the task, since the time trajectory of NF $\widetilde{X}(\tilde{t})$ still remains unknown, so (7) — is an integral equation.

To overcome this fundamental difficulty, let us apply the method that was first proposed and used by us previously to calculate the dynamics of melt−crystal and crystal−crystal fronts in the dilute metallic alloys at their fast directional solidification, and also to calculate the dynamics of amorphous material (glass) — crystal fronts at directional explosive directional explosive crystallization (EC) [32–40].

The first step of this method is the decomposition of all values included into the integrand in (7) into the Taylor's series. In particular, for the NF coordinate we have the following:

$$\widetilde{X}(\tilde{t}) - \widetilde{X}(\tilde{t}') = \widetilde{V}(\tilde{t})(\tilde{t}-\tilde{t}') - \frac{1}{2}\dot{\widetilde{V}}(\tilde{t})(\tilde{t}-\tilde{t}')^2 + \frac{1}{6}\ddot{\widetilde{V}}(\tilde{t})(\tilde{t}-\tilde{t}')^3 + \dots . \quad (9)$$

The advantage to this mathematical approach is the fact that the speed, acceleration and all hyperaccelerations of the NF trajectory in (7) become taken at one and the same fixed moment of time $\tilde{t}$ and do not depend on the alternating integration time $\tilde{t}'$. With account of (6_2) the same is related to the derivatives of the stress $\widetilde{\sigma}(\tilde{t})$ by time. Therefore, they all play a role of constant parameters and may be factored outside the integrand. The functions in the integrand are rather simple. From equations (7)−(9) it is evident that they are expressed via the gamma functions (with arguments $n = 1/2, 3/2, 5/2, \dots$), which makes it possible to calculate all integrals in the explicit form.

As a result the precise solution to the thermal conductivity equation (6_1) will be recorded in the form of an infinite series, which becomes dependent only on the stress and its derivatives by time:

$$\widetilde{T}\left(\tilde{x}=\widetilde{X}(\tilde{t}), \tilde{t}\right) - \left(1-\frac{\alpha}{\epsilon}\right) = d_0\widetilde{\sigma} + d_1\dot{\widetilde{\sigma}} + d_2\ddot{\widetilde{\sigma}} + d_3\dddot{\widetilde{\sigma}} + \dots . \quad (10)$$

Note now that if the Arrhenius equation is used (6_3), the left part (10) may be presented as the function of stress and speed. Then, using the equation (6_2), we exclude speed, so that the left part (10) becomes the function of only the stress and its first derivative by time:

$$\widetilde{T}\left(\tilde{x}=\widetilde{X}(\tilde{t}), \tilde{t}\right) - \left(1-\frac{\alpha}{\epsilon}\right) = \frac{\gamma+\alpha+\frac{1}{\epsilon}\left(1-\frac{\alpha}{\epsilon}\right)-\gamma\widetilde{\sigma}(\tilde{t})}{\epsilon-\ln\left(\widetilde{V}(\tilde{t})\right)} - \left(1-\frac{\alpha}{\epsilon}\right). \quad (11)$$

Thus (10) becomes a closed differential equation, which fully determines the stress dynamics. In its turn, its solution $\widetilde{\sigma}(\tilde{t})$ may be used in (6_2), in order to find the dependence of $\widetilde{V}(\tilde{t})$ NF speed. Then, using the known functions $\sigma(\tilde{t})$ and $\widetilde{V}(\tilde{t})$ we can find the time dependence of the temperature at NF $\widetilde{T}\left(\tilde{x}=\widetilde{X}(\tilde{t}), \tilde{t}\right)$ using Arrhenius equation (6_3), which will complete the solution of the system (6).

The success of the next, the decisive structural step, is due to the fact that in many cases there is experimental information available on the nature of the front dynamics. In particular, it is known that NF self-oscillations in process of deformation of polymer films and fibers — both in extension in process of cold drawing and in compression in process of rolling — have the nature of relaxation oscillations.

The period of relaxation oscillations consists of a short time interval, where the front speed changes quickly, and a much longer interval, where it changes smoothly, i.e. accelerations of any order become concentrated in a relatively small fraction of the period. Thus, when analyzing the self-oscillations of NF in polymers, it becomes possible to use a dimensionless small parameter — the ratio of the interval of „explosive" movement of the front to the full period of oscillations.

Sure, if the task contains a clear small dimensionless parameter, with the following proportionate to its degrees „mass" — the function that depends on speed prior to „ordinary" acceleration, and „hypermasses" — functions prior to accelerations of higher orders, the series „by accelerations" in (10) with the guarantee becomes asymptotic, and its finite section may be used for calculations with the controlled degree of precision.

Below we will show that in plastic deformation of polymers such parameter in TM (6) exists and has the following appearance:

$$\delta = \frac{2\eta}{V_s\sigma_s}\frac{(\lambda-1)}{D}. \quad (12)$$

It becomes small at high compliance of sample D and/or high speed of NF drawing V_s. It is important that both of these conditions may be met, since both the compliance degree and the drawing speed value are the control parameters in standard measurements of the plastic deformation of polymers.

Thus, we planned a workable way to build the general solution to the system (6), occurring in the self-oscillatory mode. In the next paragraph we will find out which values of dimensionless parameters introduced in paragraph 3 contribute to its rather simple implementation.

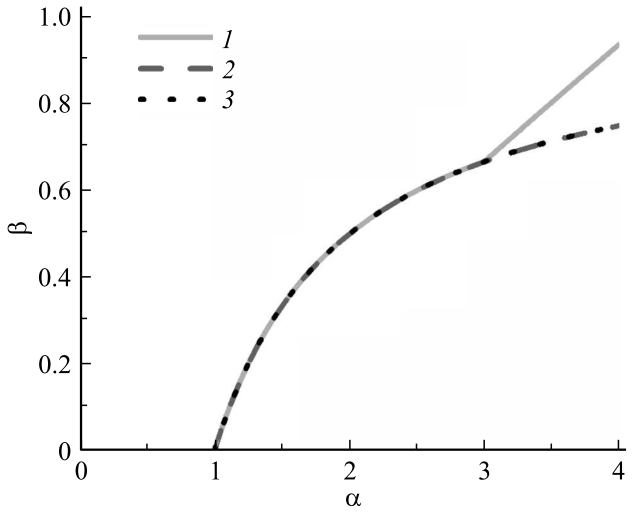


Figure 1. Diagram with lines of instability for the value of parameters $\delta = 10^{-5}$, $\gamma = 1$. Line *1* — line of instability of the system (6), determined by the exact equation (16), line *2* — line of instability of equation (19), defined by the approximate equation (22), line *3* — line of instability compliant with the Davidenkov's criterion (18).

5. Definition of the area of the parameters of existence and the value of the initial self-oscillation frequency

To solve this subtask, the method is applicable for the analysis of the linear stability of the stationary condition of the system (6). The variables of the system (6) at small deviation from the steady state may be presented as follows:

$$\widetilde{T} = 1 + \delta\widetilde{T}, \quad \widetilde{\sigma} = 1 + \delta\widetilde{\sigma},$$

$$\widetilde{X}(\tilde{t}) = \delta\widetilde{X} + \tilde{t} + 1, \quad \widetilde{V} = 1 + \delta\widetilde{V}, \tag{13}$$

where the stationary values of the neck front spread speed, temperature and stress are equal to 1. Small deviations in equations (13) will be recorded as follows:

$$\delta\widetilde{T} = r\exp(\Omega t), \quad \delta\widetilde{\sigma} = h\exp(\Omega t), \quad \delta\widetilde{V} = p\Omega\exp(\Omega t). \tag{14}$$

Substitute (13)–(14) into the system (6) and get its characteristic equation after the calculations that we added to Annex 1:

$$\Omega - \alpha\left[\frac{(\Omega+1)}{\sqrt{1+2\beta\Omega}} - 1\right] + \delta\left[\gamma + \frac{\alpha}{\sqrt{1+2\beta\Omega}}\right] = 0. \tag{15}$$

The neutral line delineating damped and undamped oscillations is specified by equation (A4):

$$\beta = \frac{\alpha M - M^2}{\alpha + \delta\gamma}, \tag{16}$$

and the values of oscillation frequencies in this line of instability are defined by the expression (A9):

$$\Omega = \frac{\sqrt{M^2 - 1}}{\alpha - M}(\alpha + \delta\gamma). \tag{17}$$

The development of the formulas (16), (17) is provided in Annex 1.

As we already wrote above, the most interesting from the experimental point of view is the case of high compliances D and/or high tensile speeds V_p. Therefore, it makes sense to first consider the case $\delta \ll 1$.

Note that at $\delta = 0$ TM (6) changes to a model that describes the dynamics of the self-sustained glass–crystal front in process of solid phase explosive EC [39]. In this case the line (17) decomposes into two lines that cross with the finite angle in the point ($\alpha = 3$, $\beta = 2/3$). However, remember that the limit $\delta = 0$ is singular, i.e. in this case, as you can easily see, the range of the system (6) is reduced.

In the area of non-zero, but small values δ, TM for polymers becomes close to the task of the forced EC, when the support with the heat from the mobile source is required for the front advancement (in practice — from the laser beam). From (15) it is evident that this analogy becomes fullest, if the frequency of oscillations is low.

Figure 1 presents the lines of instability at low value of the parameter δ. The line *1* determines the exact boundary of the system (6) self-oscillations area. If the value of parameter β is lower that $\frac{\alpha M - M^2}{\alpha + \delta\gamma}$, the system solution will be self-oscillatory.

The behavior of the frequencies for the small parameter $\delta \ll 1$ demonstrates the specific nature of the dependence on the parameter α, Figure 2. In this mode that is relevant for the dynamics of the polymers, a sharp boundary is observed in the values of the frequencies that are higher and lower than the „critical" value $\alpha \approx 3$. Therefore, in the next paragraph we will remind the corresponding experimental facts and will propose their interpretation in light of the above theoretical calculations of the oscillation frequencies.

6. About the Davidenkov's criterion

It is commonly known that the experiments for the extension of the polymer film with the specified speed of movable grip V_p measure the time dependence of stress $\sigma = \sigma(t)$. The combination of the measurement results obtained at different values V_p, makes it possible to build dependence $\sigma_s = \sigma_s(V_s)$. The self-oscillation mode is observed if such dependence has N-shaped appearance, i.e. it includes an interval where $\frac{d\sigma_s}{dV_s} < 0$.

In some experiments the existence of self-oscillations was found precisely in this interval. Therefore, their appearance was interpreted as the evident effect of compliance with the Davidenkov's criterion equivalent to meeting the condition $\frac{d\sigma_s}{dV_s} < 0$, for the occurrence of the non-stationary dynamics [41]. However, in the subsequent experiments performed with a higher resolution, it was clearly found that self-oscillations occur at speeds V_p as well in a much wider interval that it is expected from the Davidenkov's criterion. In particular, in polyethylene-terephthalate (PETP) they were observed even at the highest extension speeds, up to the maximum speed, at which the films cracked [3,19,20].

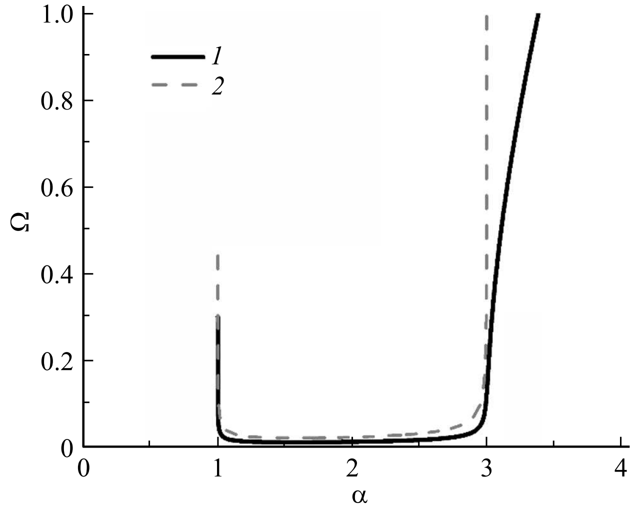


Figure 2. Dependence of the oscillation frequencies in the instability line that separates the damped and undamped oscillations. The line *1* complies with the exact formula (17), the line *2* — to the formula (21) at the parameter values $\delta = 10^{-5}$, $\gamma = 1$.

To clarify the connection to our results obtained in section 5, let us write the expression for the derivative $\frac{d\sigma_s}{dV_s}$ in dimensionless parameters:

$$\frac{d\ln\sigma_s}{d\ln V_s} = \frac{1-\alpha(1-\beta)}{\alpha+\gamma}. \tag{18}$$

From equation (18) it follows that the Davidenkov's criterion $\frac{d\sigma_s}{dV_s} < 0$ corresponds to the condition $\beta < \frac{\alpha-1}{\alpha}$. Having compared it with the lines of instability for the initial system (6), we can see that the negative derivative in (18) corresponds to the part of the line of instability at $1 < \alpha < 3$, $\delta \to 0$, Figure 1. I.e. in this segment the Davidenkov's criterion is very close to the results obtained for small δ. However, at $\alpha > 3$ the line of instability of the system (6) „turns left" from the line $\beta = \frac{\alpha-1}{\alpha}$, and the sign of the derivative in (18) in this segment becomes positive. Accordingly, in this part of the line of instability, despite the fact that when it is crossed „to the right", i.e. to the large values α, the oscillations arise, and the Davidenkov's criterion is not met. High-frequency oscillations at $\alpha \geq 3$ or $\beta \geq 2/3$ correspond to high extension speeds. It is exactly for the high extension speeds that the Davidenkov's criterion will not predict the oscillations that are observed experimentally.

Besides, the results of this analysis make it possible to understand why in some experiments the machine stopped recording the oscillations upon achievement of the maximum speed predicted by the Davidenkov's criterion (even though it sometimes recorded them a bit higher than this value [23]). There are two reasons here: first, a very sharp growth of the frequency already when this speed is somewhat exceeded, Figure 2. The second cause — is the decrease of the dimensions of the stable limit cycle with the speed increase. If the experimental setup resolution is not high enough, these both causes made it impossible to find the presence of the high frequency self-oscillations at high speeds, but recorded them at low speeds, when both the oscillation frequencies are „convenient", and their oscillation amplitudes are maximum.

Qualitatively one may say that the dynamics of NF (and also of the stress and temperature) disintegrates into two different modes, „mechanical" (Davidenkov's) — with small frequencies and large amplitudes and „thermodynamic" — with high frequencies and small amplitudes. We will discuss the principal difference in the classification of the mechanisms that each of these modes is based on in the final part of the article.

From the given results it is clear that the analysis of the non-linear dynamics of NF must be carried out differently for areas $1 < \alpha < 3$ and $\alpha > 3$. Besides, the program planned in section 4 for the approximate non-linear differential equation is easiest to be completed in the first region, where the frequency of self-oscillations is expected to be low (at least at shallow advancement beyond the line of instability), and the series mentioned in section 4 „by accelerations" in equation (10) is asymptotic and has the evident small parameter at $\delta \ll 1$.

Considering that from the experimental point of view as well the case of small values δ is of great interest, in the next paragraph we will develop a differential equation that describes the non-linear dynamics in the area $1 < \alpha < 3$ and will analyze its solutions.

7. Solution to task (1)–(3) in the approximation of the non-linear oscillator

In the area of the small values of the equation (17) frequencies you can „break" the series in (10), leaving only the contributions of not higher than the 2nd derivative by time. As a result, using equations (6_2), (6_3), (7)–(9), after the corresponding calculations given in Annex 2, we get the ordinary non-linear differential equation for the deviation of the dimensionless stress from its stationary value $h(t) \equiv \tilde{\sigma}(t) - 1$ in the form of:

$$M(h,\dot{h})\ddot{h} + F(h,\dot{h}) + \delta h = 0, \tag{19}$$

where the effective mass and friction force are equal to:

$$M(h,\dot{h}) = \frac{\alpha}{8\gamma}\frac{\epsilon - \ln(\tilde{V})}{\epsilon}\beta_0(\tilde{V})\Big\{(h+1)\big[2 - 3\beta_0(\tilde{V})(\tilde{V})^2\big] + \frac{3}{2}\beta_0(\tilde{V})\Big[2\delta + \dot{h}\big(-3 + 5\beta_0(\tilde{V})(\tilde{V})^2\big)\Big]\Big\},$$

$$F(h,\dot{h}) = \frac{\alpha\delta}{\gamma}\left(\frac{\beta_0(\tilde{V})}{\beta}\right)^{1/2}\frac{\epsilon - \ln(\tilde{V})}{\epsilon}\Big\{-\frac{1}{2}\beta_0(\tilde{V})\tilde{V}\dot{h} + \tilde{V}(h+1) - \sqrt{\frac{\beta}{\beta_0(\tilde{V})}}\Big\} - \frac{\delta}{\gamma}\ln(\tilde{V}). \tag{20}$$

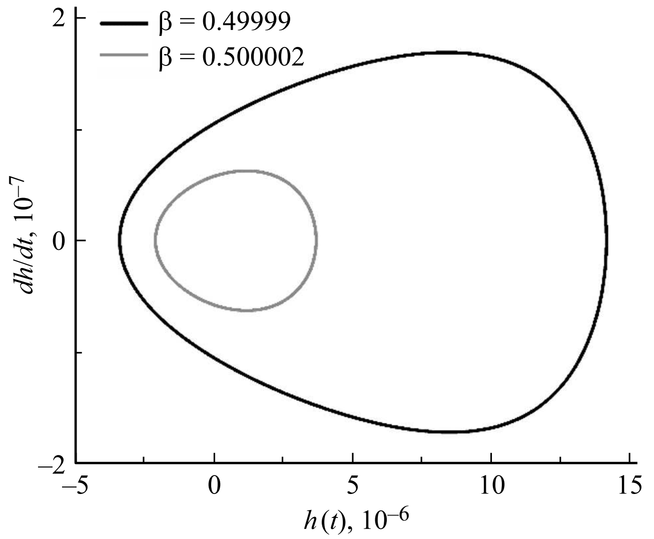


Figure 3. Phase diagrams of equation (19) at values of parameters $\delta = 10^{-5}$, $\gamma = 1$, $\epsilon = 10$.

In this equation „the friction force" $F(h, \dot{h})$ has non-monotonic dependence. Note that at small δ its linear part is

$$\frac{dF}{d\dot{h}}\bigg|_{h=\dot{h}=0} = \frac{d\sigma_s}{dV_s} + O(\delta),$$

which provides for the occurrence of the stable limit cycle in the phase plane (α, β). It is clear therefore that the self-oscillations provided for by the presence of the limit cycle, already at the relatively small advancement into the area of instability, shall assume the character of relaxation oscillations.

Linearization of equation (19) makes it possible to obtain the estimate of the oscillation frequency Ω:

$$\Omega = \sqrt{8\delta(1-\delta)\,\frac{\alpha}{\alpha-1}\,\frac{\gamma+\alpha}{3-\alpha}}, \tag{21}$$

in the corresponding line of instability:

$$\beta = \frac{\alpha - 1}{\alpha(1-\delta)}. \tag{22}$$

Figure 3 shows the considerable increase of the cycles in the phase plane when advancing into the area of self-oscillations. Figure 4, *a, b* shows the corresponding time dependences of temperature and stress deviations from their stationary values.

From the curves in Figures 3–4 it is evident that the solutions to the equation (19) stop being sinusoidal and become more relaxational further from the line of instability (22).

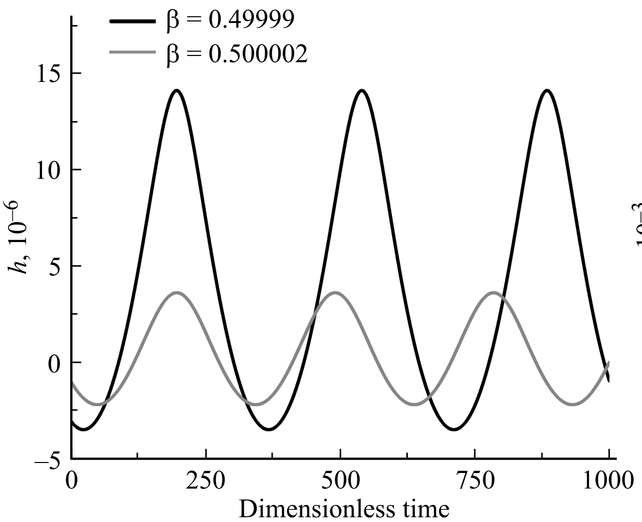
8. Conclusion

Having reformulated the thermal conductivity equation for the polymer film extended with the constant speed, in the form of a differential equation of infinite order „by accelerations", we were able to ask the question: ≪is it possible under certain conditions to obtain the good approximation of self-oscillatory solutions to the initial task „by cutting the series", i.e. solving the differential equation of the finite order?≫.

Besides, we, first of all, were interested in a situation when it is necessary to increase the system compliance to initiate the self-oscillations, decreasing the parameter δ. This is true for most studied polymers, where self-oscillations were observed.

Having identified a rather small number of dimensionless parameters from the multiple size parameters of the model, we demonstrated that within the limit of the infinitely high compliance „the polymer" model reduces to the solution of a much simpler model of spontaneous solid-phase explosive solidification of glasses.

In the real case, then the compliance is not infinite, but high (and/or the film cold drawing speed is high), the



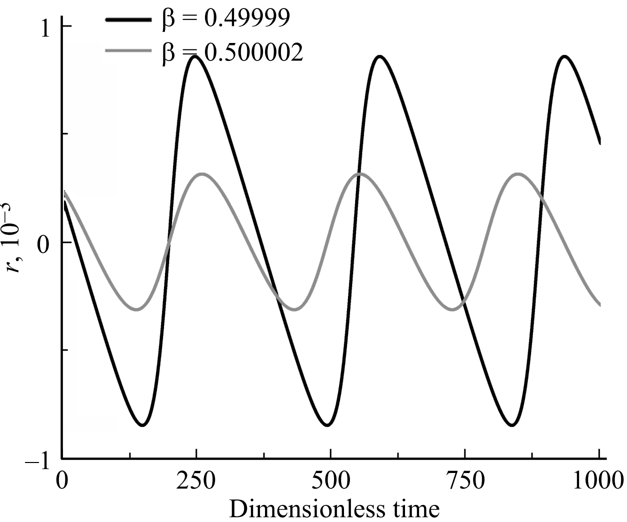


Figure 4. Dependence in self-oscillatory mode *a*) between stress and time $h(\tilde{t})$, *b*) temperature and time $r(\tilde{t})$.

calculation of the „polymer" dynamics becomes equivalent to the solution of the task on the advancement of the inorganic glass-crystal front induced by the laser beam „pushing" it. At the same time it was rather unexpected that the reverse compliance of the film plays the same role as the temperature gradient from the laser backlighting [39].

Note that both these analogies are interesting as such, since they enable the comparison of the known results in two totally different at first sight types of experiments.

The results of the analysis of the polymer film linear stability in the article, where the film moves with the same (constant) speed ad the movable grip, fully agree with this analogy. Besides, it was found that at not so intense heating of the neck front, the frequency of the arising oscillations is low, which makes it possible to reasonably approximate the calculation of the film dynamics by solving of an ordinary differential equation of the 2nd order, i. e. the non-linear oscillator.

Whereas in the case of intense heating, the self-oscillation frequency increases sharply, and the size of the stable limit cycle decreases. This pattern shows clearly in the change of the time dependence of stress on drawing speed observed experimentally.

Finally, in connection with the general form of the developed equation (10), it is appropriate to pay attention to the fundamental distinctive feature in the description of self-oscillation dynamics in the polymers from self-oscillations in some macroscopic models widely used in mechanics, electrical engineering, and chemistry. Indeed, the latter are described by differential equations of the 2nd order (classic Van-der-Pol and Rayleigh with the permanent coefficients or more general equations (such as Lienard equations), containing the variable coefficients). For the unambiguous definition of their solutions, it is possible to specify only 2 initial values. At the same time the presentation of the thermal conductivity equation (1) in the form of a differential equation of infinite order (10) to determine the trajectory requires setting the infinite number of the initial values. It is evident that this circumstance may potential cause the occurrence of the non-linear dynamics, which is much more complex than self-oscillations „of ordinary" macromodel.

Physically speaking, the difference in „classification of mechanisms" of self-oscillations (predictable in [19]) in polymers and simple macromechanical models (such as the mechanical clock model) consists in the infinite or finite number of degrees of freedom that is the minimum necessary for reasonable specification of the model equations. From here the cause is intuitively clear for the increase of the period of self-oscillations the integer number of times with the very small change in the control parameters, which was observed many times in deformation of polymers [3,14] — because it is „much more difficult" for the dynamic state of the neck front to return for the period to the same point in the infinite phase space compared to the same for the point in the plane, therefore „the polymer" trajectory has to make several „nearly successful" attempts before it finally hits the „mark" and the period time expires.

Therefore, the results obtained in the article made it possible to also name the common cause for the occurrence of self-oscillations in the polymers as in the relatively narrow area of compliance with the mechanical Davidenkov's criterion and their sudden appearance in a much wider area of the speeds, where this criterion is not met.

The approach specified in the article may be used for the calculation of the non-stationary dynamics of polymer films with account of the increase in the free volume and the degree of drawing arising with the transition of the neck temperature through the glass transition temperature. In virtue of the substantial complication of the formulas such calculations will be provided in the next publication.

Annex 1

To solve the system (6) it is especially interesting to solve the characteristic equation (15) with purely imaginary roots $\Omega = i\Omega_{im}$. Let's represent the (15) for such roots in the following form:

$$\sqrt{1+2\beta i\Omega_{im}} = \frac{\alpha(i\Omega_{im}+1-\delta)}{i\Omega_{im}+(\alpha+\delta\gamma)}.$$

The value of the left and right parts will be recorded as complex numbers:

$$\sqrt{1+2\beta i\Omega_{im}} = \frac{\alpha(i\Omega_{im}+1-\delta)}{i\Omega_{im}+(\alpha+\delta\gamma)} = M + iN,$$

where M and N are real numbers.

From the condition $\sqrt{1+2\beta i\Omega_{im}} = M + iN$ it follows that

$$N^2 = M^2 - 1, \quad \Omega_{im} = \frac{MN}{\beta_p}. \tag{A1}$$

Since M and $N \in R$ then $|M| \geq 1$follows.

From the condition

$$\frac{\alpha(i\Omega_{im}+1-\delta_p)}{i\Omega_{im}+(\alpha+\delta\gamma)} = M + iN,$$

it follows that

$$\begin{cases} \alpha\Omega_{im} = M\Omega_{im} + N(\alpha+\delta\gamma), \\ \alpha(1-\delta) = M(\alpha+\delta\gamma) - N\Omega_{im}. \end{cases}$$

Let us use the equations (A1), and this system will look as follows:

$$\begin{cases} \alpha\dfrac{M}{\beta} = \dfrac{M^2}{\beta} + (\alpha+\delta\gamma), \\ \alpha(1-\delta) = M(\alpha+\delta\gamma) - \dfrac{M(M^2-1)}{\beta}. \end{cases} \tag{A2}$$

Having excluded β from the equations of the system (A2), we will get the condition in M via the parameters

α, δ and γ:

$$2(\alpha+\delta\gamma)M^2 - \alpha[(\alpha+\delta\gamma)+(1-\delta)]M + [\alpha^2(1-\delta) - (\alpha+\delta\gamma)] = 0.$$

Let us write the explicit expression for M from parameters α, δ, γ as the solution to the square equation:

$$M = \frac{\alpha[(1-\delta)+(\alpha+\delta\gamma)] \pm \sqrt{D_M}}{4(\alpha+\delta\gamma)}, \qquad \text{(A3)}$$

where

$$D_M = \alpha^2[(1-\delta)+(\alpha+\delta\gamma)]^2 - 8(\alpha+\delta\gamma)[\alpha^2(1-\delta)-(\alpha+\delta\gamma)],$$

The corresponding value of the frequency according to (A1) is:

$$\Omega_{im} = \frac{M\sqrt{M^2-1}}{\beta}. \qquad \text{(A4)}$$

Let us write the expression for the parameter β from the system (A2)

$$\beta = \frac{\alpha M - M^2}{\alpha+\delta\gamma}. \qquad \text{(A5)}$$

The ratio (A5) defines the line of instability separating damped and non-damped oscillations. The value of frequency (A4) at values β (A5) is:

$$\Omega_{im} = \frac{\sqrt{M^2-1}}{\alpha - M}(\alpha+\delta\gamma). \qquad \text{(A6)}$$

Annex 2

The solution to the thermal conductivity equation (6_1) will be written via the Green's function:

$$\widetilde T\big(\widetilde X(\tilde t), \tilde t\big) - \left(1-\frac{\alpha}{\epsilon}\right) = \frac{\alpha}{\epsilon\sqrt{\beta}} \int_{-\infty}^{\tilde t} dt' \frac{1}{\sqrt{\pi(\tilde t - \tilde t')}} \times \exp\left[-\frac{1-\beta}{\beta}(\tilde t - \tilde t') - \frac{[\widetilde X(\tilde t) - \widetilde X(\tilde t')]^2}{(\tilde t - \tilde t')}\right] \times \widetilde\sigma\Big(\widetilde V(\tilde t'), \widetilde T\big(\widetilde X(\tilde t')\big)\Big)\widetilde V(\tilde t'). \qquad \text{(A7)}$$

Let us write the functions $\widetilde X(\tilde t)$, $\widetilde\sigma(\tilde t)$, $\widetilde V(\tilde t')$ in the form of Taylor's series:

$$\widetilde X(\tilde t') = \widetilde X(\tilde t) - \widetilde V(\tilde t)\tilde\tau + \frac{1}{2}\dot{\widetilde V}(\tilde t)\tilde\tau^2 - \frac{1}{6}\ddot{\widetilde V}(\tilde t)\tilde\tau^3 + \dots$$

$$\widetilde\sigma(\tilde t') = \widetilde\sigma(\tilde t) - \dot{\widetilde\sigma}(\tilde t)\tilde\tau + \frac{1}{2}\ddot{\widetilde\sigma}(\tilde t)\tilde\tau^2 - \frac{1}{6}\dddot{\widetilde\sigma}(\tilde t)\tilde\tau^3 + \dots, \qquad \text{(A8)}$$

where $\tilde\tau = \tilde t - \tilde t'$.

Let us substitute the contributions that are substantial at low frequencies and written in equations (A8), to the right part (A7) and get the following ratio:

$$\widetilde T - \left(1 - \frac{\alpha}{\epsilon}\right) = \widetilde\sigma I_1 + I_2, \qquad \text{(A9)}$$

where

$$I_1 = \frac{\alpha}{\epsilon\sqrt{\beta}} \int_0^\infty d\tilde\tau \frac{1}{\sqrt{\pi\tilde\tau}} f_1(\tilde\tau), \qquad \text{(A10)}$$

$$f_1(\tilde\tau) = \exp\left[-\frac{1-\beta}{\beta}\tilde\tau - \tilde\tau\left[\widetilde V - \frac{1}{2}\dot{\widetilde V}\tilde\tau\right]^2\right][\widetilde V - \dot{\widetilde V}\tilde\tau],$$

$$I_2 = \frac{\alpha}{\epsilon\sqrt{\beta}} \int_0^\infty d\tilde\tau \frac{1}{\sqrt{\pi\tilde\tau}} f_2(\tilde\tau), \qquad \text{(A11)}$$

$$f_2(\tilde\tau) = \exp\left[-\frac{1-\beta}{\beta}\tilde\tau - \tilde\tau\left[\widetilde V - \frac{1}{2}\dot{\widetilde V}\tilde\tau\right]^2\right] \times \left[-\dot{\widetilde\sigma}\tilde\tau + \frac{1}{2}\ddot{\widetilde\sigma}\tilde\tau^2\right][\widetilde V - \dot{\widetilde V}\tilde\tau],$$

where $\tilde t$ — argument in the sought functions $\widetilde\sigma$, $\widetilde V$ and their derivatives, it is omitted for brevity of the record. Let's represent the exponent in the integrand as follows:

$$\exp\left[-\left(\frac{1-\beta}{\beta} + \widetilde V^2\right)\tilde\tau\right]\left[1 + \widetilde V\dot{\widetilde V}\tilde\tau^2 - \frac{1}{4}\dot{\widetilde V}^2\tilde\tau^3\right], \qquad \text{(A12)}$$

where the insignificant contributions were omitted because of the low value of oscillation frequency.

The functions $f_1(\tilde\tau)$ and $f_1(\tilde\tau)$ in accordance with (A12) will look like:

$$f_1(\tilde\tau) = \exp\left[-\left(\frac{1-\beta}{\beta} + \widetilde V^2\right)\tilde\tau\right] \times \left[1 + \widetilde V\dot{\widetilde V}\tilde\tau^2 - \frac{1}{4}\dot{\widetilde V}^2\tilde\tau^3\right][\widetilde V - \dot{\widetilde V}\tilde\tau]. \qquad \text{(A13)}$$

$$f_2(\tilde\tau) = \exp\left[-\left(\frac{1-\beta}{\beta} + \widetilde V^2\right)\tilde\tau\right]\left[1 + \widetilde V\dot{\widetilde V}\tilde\tau^2 - \frac{1}{4}\dot{\widetilde V}^2\tilde\tau^3\right] \times \left[-\dot{\widetilde\sigma}\tilde\tau + \frac{1}{2}\ddot{\widetilde\sigma}\tilde\tau^2\right][\widetilde V - \dot{\widetilde V}\tilde\tau]. \qquad \text{(A14)}$$

Let us open the brackets in (A13), (A14), imagine $f_1(\tilde\tau)$ and $f_2(\tilde\tau)$ as the products of the polynomials by degrees $\tilde\tau$ per exponent

$$\exp\left[-\left(\frac{1-\beta}{\beta} + \widetilde V^2\right)\tilde\tau\right].$$

Besides, in (A13), (A14) we will leave only those members that contain the non-linear members relative to the derivatives of not higher than the second degree by time and the linear members including the second derivative. The cause for such simplification is the fact that in this article we study the mode with low frequencies of self-oscillations

$$f_1(\tilde{\tau}) = (\tilde{V} - \dot{\tilde{V}}\tilde{\tau} + \tilde{V}^2\dot{\tilde{V}}\tilde{\tau}^2)\exp\left[-\left(\frac{1-\beta}{\beta} + \tilde{V}^2\right)\tilde{\tau}\right], \quad (A15)$$

$$f_2(\tilde{\tau}) = \left[-\tilde{V}\dot{\tilde{\sigma}}\tilde{\tau} + \left(\dot{\tilde{V}}\dot{\tilde{\sigma}} + \frac{1}{2}\ddot{\tilde{\sigma}}\tilde{V}\right)\tilde{\tau}^2 - (\tilde{V}^2\dot{\tilde{V}}\dot{\tilde{\sigma}})\tilde{\tau}^3\right] \times \exp\left[-\left(\frac{1-\beta}{\beta} + \tilde{V}^2\right)\tilde{\tau}\right]. \quad (A16)$$

Integrals $I_1(\tilde{t})$ and $I_2(\tilde{t})$ (A10), (A11) with account of (A15)–(A16) are calculated easily, since they are the sums of the gamma functions. The value $I_1(\tilde{t})$ and $I_2(\tilde{t})$ are equal to:

$$I_1(\tilde{t}) = \frac{\alpha}{\epsilon\sqrt{\beta}}\left(\beta_0^{1/2}\tilde{V} - \frac{1}{2}\beta_0^{3/2}\dot{\tilde{V}} + \frac{3}{4}\beta_0^{5/2}\tilde{V}^2\dot{\tilde{V}}\right),$$

$$I_2(\tilde{t}) = \frac{\alpha}{\epsilon\sqrt{\beta}}\left(-\frac{1}{2}\beta_0^{3/2}\tilde{V}\dot{\tilde{\sigma}} + \frac{3}{4}\beta_0^{5/2}\left(\dot{\tilde{V}}\dot{\tilde{\sigma}} + \frac{1}{2}\ddot{\tilde{\sigma}}\tilde{V}\right) - \frac{15}{8}\beta_0^{7/2}\tilde{V}^2\dot{\tilde{V}}\dot{\tilde{\sigma}}\right),$$

where the function

$$\beta_0 = \beta_0(\tilde{V}) = \frac{1}{\frac{1-\beta}{\beta} + \tilde{V}^2}.$$

Imagine I_1 and I_2 in the form of the function of deviation from the stationary value of stress $h(\tilde{t}) = \tilde{\sigma}(\tilde{t}) - 1$, with account of ($6_2$) I_1 and I_2 will look like:

$$I_1(h, \dot{h}, \ddot{h}) = \frac{\alpha}{\epsilon\sqrt{\beta}}\left(\beta_0^{1/2}\tilde{V} + \frac{1}{4\delta}\beta_0^{3/2}\ddot{h} - \frac{3}{8\delta}\beta_0^{5/2}\tilde{V}^2\ddot{h}\right),$$

$$I_2(h, \dot{h}, \ddot{h}) = \frac{\alpha}{\epsilon\sqrt{\beta}}\left(-\frac{1}{2}\beta_0^{3/2}\tilde{V}\dot{h} + \frac{3}{4}\beta_0^{5/2}\left(-\frac{1}{2\delta}\dot{h}\ddot{h} + \frac{1}{2}\ddot{h}\tilde{V}\right) - \frac{15}{16\delta}\beta_0^{7/2}\tilde{V}^2\ddot{h}\dot{h}\right),$$

where $\tilde{V} = 1 - \frac{1}{2\delta}\dot{h}$.

Then the right part of the equation (A9) depends only on the unknown function h:

$$\tilde{T} - \left(1 - \frac{\alpha}{\epsilon}\right) = (h+1)I_1(h, \dot{h}, \ddot{h}) + I_2(h, \dot{h}, \ddot{h}). \quad (A17)$$

The left part of the ratio (A9) $\tilde{T} - \left(1 - \frac{\alpha}{\epsilon}\right)$ will be expressed from the equation $(6+3)$:

$$\tilde{T}(\tilde{X}(\tilde{t}), \tilde{t}) - \left(1 - \frac{\alpha}{\epsilon}\right) = \frac{\gamma h - \ln(\tilde{V})}{\ln(\tilde{V}) - \epsilon} + \frac{\alpha}{\epsilon}. \quad (A18)$$

Having combined (A17) and (A18), we will get the differential equation at h:

$$\frac{\gamma h - \ln(\tilde{V})}{\ln(\tilde{V}) - \epsilon} + \frac{\alpha}{\epsilon} = (h+1)I_1(h, \dot{h}, \ddot{h}) + I_2(h, \dot{h}, \ddot{h}). \quad (A19)$$

The equation (A19), which is an ordinary differential equation of the second order, may be written briefly in the form of an equation for a non-linear oscillator:

$$M(h, \dot{h})\ddot{h} + F(h, \dot{h}) + \delta h = 0, \quad (A20)$$

where the effective mass and friction force are equal to:

$$M(h, \dot{h}) = \frac{\alpha\beta_0}{8\gamma}\sqrt{\frac{\beta_0}{\beta}}\frac{\epsilon - \ln(\tilde{V})}{\epsilon} \times \left[(h+1)(2 - 3\beta_0\tilde{V}^2) + \frac{3}{2}\beta_0\left(2\delta + (-3 + 5\beta_0^2\tilde{V}^2)\dot{h}\right)\right],$$

$$F(h, \dot{h}) = \frac{\alpha\delta}{\gamma}\sqrt{\frac{\beta_0}{\beta}}\frac{\epsilon - \ln(\tilde{V})}{\epsilon} \times \left[(h+1)\tilde{V} - \frac{1}{2}\beta_0\tilde{V}\dot{h} - \sqrt{\frac{\beta_0}{\beta}}\right] - \frac{\delta}{\gamma}\ln(\tilde{V}).$$

The solution to the equation (A20) defines the dimensionless stress $\tilde{\sigma} = h + 1$, the speed of the neck front $\tilde{V} = 1 - \frac{1}{2\delta}\dot{h}$ and the temperature at the neck front:

$$\tilde{T} - 1 = \frac{\alpha}{\epsilon}\left[-1 + \sqrt{\frac{\beta_0}{\beta}}\left[(h+1)\left(\tilde{V} + \frac{\beta_0}{8\delta}(2 - 3\beta_0\tilde{V}^2)\ddot{h}\right) + \beta_0\left(-\frac{1}{2}\tilde{V}\dot{h} + \frac{3\beta_0}{16\delta}\left(2\delta + (-3 + 5\beta_0\tilde{V}^2)\dot{h}\right)\ddot{h}\right)\right]\right]. \quad (A21)$$

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Conflict of interest

The authors declare that they have no conflict of interest.

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