# PHYSICAL CHEMISTRY

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1963

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**Abstract**

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# ON CRITERIA FOR THE INSTABILITY OF REGIMES OF SHEAR DEFORMATION OF ELASTIC-VISCOUS POLYMER SYSTEMS

*(Presented by Academician V. A. Kargin on 4 February 1963)*

The deformation of polymer systems is accompanied by a number of phenomena to which special attention has recently been drawn in connection with the process of flow of polymer melts at high shear stresses. As an illustration one may cite photographs, obtained by N. V. Prozorovskaya in our institute, of jets of polymers flowing out of cylindrical capillaries (Fig. 1). Phenomena analogous to those presented in Fig. 1 have been described more than once in the literature ([1–11]). In all such cases, upon reaching certain critical values of the parameters that determine the deformation regimes, the polymer jet issuing from the dies acquires regular or irregular disturbances: it becomes screw-like, constrictions appear on it, etc. At very high shear stresses and shear rates, polymer jets may break up into separate grains of irregular shape. Disturbances in the flow of polymers play a very important role in rotational (disk) extruders and in many other cases. Attempts have been made ([1]) to relate the onset of an unstable regime in polymer systems to the magnitude of the Reynolds criterion. However, in work ([2]) the fundamental inadmissibility of such an approach was convincingly demonstrated. A number of authors ([3–5]) believe that the onset of flow irregularity is due to the high elasticity of polymer systems. In ([2,6–8]) the influence of the entrance and exit effects on the onset of irregularities during the flow of polymers in capillaries was studied. There is also an opinion ([3]) that irregularities arise within the capillary itself. Many authors ([1,4,8–10]) considered the influence of temperature and molecular weight of the specimens on the critical shear stresses. In work ([5]) it is indicated that the onset of irregularities is associated with a definite ratio between the tangential and normal stresses in the melt. It was found ([4]) that the magnitude of the elastic shear deformation at the onset of irregular flow reaches a definite value and remains substantially constant for different materials.

**Fig. 1.** Samples of polymer jets (polyethylene and polypropylene) flowing from a capillary at different shear rates and shear stresses: **1** –steady jet; **2** –beginning of an unstable regime; **3** –formation of a regular spiral; **4** –jet that has broken up into separate grains.

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In the present work the phenomenon of flow instability and the manifestation of disturbances are considered by methods of the theory of similarity. In this way a system of criteria is obtained that characterizes the process of deformation of elastic-viscous media.

Let us first consider a one-dimensional plane deformation of an elastic-viscous medium obeying the well-known Maxwell equation

$$\frac{\partial V}{\partial t} + \theta \frac{\partial^2 V}{\partial t^2} = \nu \frac{\partial^2 V}{\partial y^2}, \tag{1}$$

here $V$ is the velocity of motion, depending on the time $t$ and on the coordinate $y$, directed across the flow; $\nu$ is the kinematic viscosity of the system, and $\theta$ is the relaxation time.

Let now $r$, $U$, and $T$ be, respectively, the characteristic length, velocity, and time defining the process.

Passing to dimensionless variables $x = y/r$, $u = V/U$, $\tau = t/T$, and multiplying all terms of the equation by the quantity $r/U^2$, we obtain

$$\frac{r}{UT}\frac{\partial u}{\partial \tau} + \frac{\theta r}{UT^2}\frac{\partial^2 u}{\partial \tau^2} = \frac{\nu}{Ur}\frac{\partial^2 u}{\partial x^2}. \tag{2}$$

If the second term on the left-hand side of equation (2) is small in comparison with the first, then (2) becomes the equation of flow of a viscous liquid; if, conversely, the first term on the left-hand side of this equation is small, then the equation of elastic oscillations is obtained. The complexes standing before the derivatives are dimensionless criteria of similarity:

$$\frac{\nu}{Ur} = \frac{1}{\mathrm{Re}}, \qquad \frac{r}{UT} = \frac{1}{\mathrm{Ho}}, \qquad \frac{\theta r}{UT^2} = \mathrm{Re}_e,$$

where Re is the Reynolds criterion, and Ho is the homochronicity criterion. The quantity $\mathrm{Re}_e$ is a new criterion, which we shall call the elastic Reynolds criterion. Consequently, in the case of deformation of elastic-viscous media, in addition to the usual criteria Re, Ho, the flow dynamics is also determined by the criterion $\mathrm{Re}_e$.

If equation (1) is replaced by Kelvin' s law of deformation of a body, and then dimensionless variables are introduced and the similarity criteria are isolated, the same quantities are obtained, namely Re, Ho, $\mathrm{Re}_e$. One may also use the Kartin–Slonimskii model, which well describes the properties of real polymer systems [(12)]. In this case as well a system of criteria is obtained, some of which characterize the elastic-viscous properties of the system.

The criterion $\mathrm{Re}_e$ appears because of the presence of elastic forces in the material. It is a measure of the ratio of viscous forces to elastic forces. Upon reaching a certain critical $\mathrm{Re}_e$, the elastic oscillations in the moving medium can no longer be damped by its viscosity, i.e., the flow ceases to be stable with respect to any small perturbations of the stream. The criterion $\mathrm{Re}_e$ may be regarded as an analogue of the ordinary criterion Re. Therefore the expression "elastic turbulence," encountered in the literature [(4)] to describe the phenomenon under study, has by chance proved to be very apt.

Since the manifestation of elastic properties in the deformation of elastic-viscous bodies depends on the rate of deformation and, consequently, on the distribution of velocities in the direction transverse to the direction of deformation, it is natural to relate the characteristic time to the relative velocity, i.e., to the shear rate $\dot{\gamma}$; then $T = 1/\dot{\gamma}$.

A generalization of the viscoelastic liquid considered by us is a medium whose deformation law is described by the following equation:

$$a_1 \frac{\partial V}{\partial y} + a_2 \frac{\partial}{\partial t}\left(\frac{\partial V}{\partial y}\right) + ... + \alpha_{n+1} \frac{\partial^n}{\partial t^n}\left(\frac{\partial V}{\partial y}\right) =$$

$$= \beta_1 p + \beta_2 \frac{\partial p}{\partial t} + ... + \beta_{m+1} \frac{\partial^m p}{\partial t^m}, \tag{3}$$

where $\alpha_i$ and $\beta_i$ are empirical constants characterizing the individual properties of the medium.

Proceeding in the same way as was done above for a Maxwell liquid, one can obtain:

$$\frac{\partial^2}{\partial x^2} \sum_{k=0}^{n} B_k \frac{\partial^k u}{\partial \tau^k} = \frac{\partial}{\partial t} \sum_{k=0}^{n} A_k \frac{\partial^k u}{\partial \tau^k}, \tag{4}$$

where

$$A_k = \frac{\beta_{k+1}}{\beta_1 T^k} \frac{1}{H_0}, \qquad B_k = \frac{\alpha_{k+1}}{\alpha_1 T^k} \frac{1}{\mathrm{Re}}.$$

a set of criteria characterizing the deformation of the medium described by equation (3).

If only the simplest elastic-viscous liquid is considered, then, besides Ho and Re, there appears the criterion $A_1$, which was previously denoted by $\mathrm{Re}_e$.

The use of the complete set of criteria makes it possible to solve various problems arising in the flow of viscous-elastic liquids. However, the application of only the first criterion, namely $\mathrm{Re}_e$, already makes it possible to explain qualitatively, and sometimes quantitatively, the set of observations associated with elastic turbulence arising when polymers flow out of nozzles.

For the case of flow in capillaries, the quantities entering the criterion $\mathrm{Re}_e$ acquire the following meaning: $r$ is the radius of the capillary, $U$ is the mean velocity of flow of the medium. As was assumed earlier, $T = 1/\dot{\gamma}$; then the following expressions for $\mathrm{Re}_e$ may be written:

$$\mathrm{Re}_e = \frac{\theta}{T} = \theta\dot{\gamma} = \gamma, \tag{5}$$

where $\gamma$ is the reversible elastic deformation of the polymer system. If it is assumed [13] that the normal stresses $\sigma$ are related to $\gamma$ by the equation

$$\sigma = G\gamma^2, \tag{6}$$

then

$$\mathrm{Re}_e = \frac{\tau}{\sigma}. \tag{7}$$

Here the shear stresses arise as a result of the viscosity of the system, and the normal stresses as a consequence of its elasticity.

It is seen from (5) that, in its physical meaning, $\mathrm{Re}_e$ is the ratio of the relaxation time to the duration of the process. This is in complete agreement with modern ideas on the mechanism of deformation of polymer systems [12].

The critical value $\mathrm{Re}_{\mathrm{ek}}$, upon attainment of which elastic turbulence sets in, follows from the preceding reasoning to be a universal constant independent of temperature, the geometrical dimensions of the capillary, and the nature of the materials under investigation.

Let us consider the dependence of the rate of flow through a capillary at the onset of the critical flow regime ($Q_\mathrm{k}$) on the viscosity of the material $\eta$ and on the capillary radius $r$. The mean effective shear rate is $\dot{\gamma} = D = Q/\pi r^3$; therefore:

$$\mathrm{Re}_{\mathrm{ek}} = \frac{Q_\mathrm{k}\eta}{\pi r^3 G}. \tag{8}$$

It follows from this that $Q_\mathrm{k}$ is directly proportional to the cube of the radius and inversely proportional to the viscosity. These conclusions correspond to the experimental data of many authors analyzed by Tordella [(2)].

Let us consider the dependence of the critical shear rates $\dot{\gamma}_\mathrm{k}$ and shear stresses $\tau_\mathrm{k}$ on the temperature $T$. From (5) it follows that

$$\dot{\gamma}_\mathrm{k}(T) = \mathrm{Re}_{e\mathrm{k}} \frac{GT(T)}{\eta(T)}. \tag{9}$$

The dependence of the modulus $G$ on temperature is weak (for the temperature range under consideration); therefore $\dot{\gamma}_\mathrm{k}(T) \sim 1/\eta(T)$. Since the viscosity decreases rather sharply with temperature, $\dot{\gamma}_\mathrm{k}$ should increase rapidly. This agrees well with all available experimental data [(2,4)].

The dependence of $\tau_\mathrm{k}$ on temperature is weakly expressed, since $\tau_\mathrm{k} \sim G(T)$. From the molecular-kinetic theory of high elasticity [(14)] it follows that $G \sim \rho T$, where $\rho$ is the density of the material. The quantity $\rho T$ usually increases slowly with temperature. Therefore $\tau_\mathrm{k}$ should increase slightly as the temperature rises. This fact has been observed by a number of investigators [(2,4)]. When considering the dependence $\tau_\mathrm{k}(T)$, it should be borne in mind that the experimental determination of $\tau_\mathrm{k}$ is associated with the possibility of considerable errors.

([2,10]), and deviations from the relation $\tau_k = \mathrm{const}$ lie, as a rule, within the limits of experimental error.

Taking (14), namely that $G \sim \dfrac{RT\rho}{M}$, we obtain

$$\tau_k \sim \frac{RT\rho}{M} \mathrm{Re}_{ek}, \tag{10}$$

where $R$ is the gas constant. Thus, at a given temperature, $\tau_k$ is inversely proportional to the molecular weight, which has been observed experimentally ([4,8]).

Metzner ([9]) calculated the ratio $\tau/\sigma$ for two polymers under the critical flow regime. However, $\sigma$ was not calculated by formula (7), and therefore the critical values $(\tau/\sigma)_k$ proved to be different for the two materials. As has become clear, the hypothesis advanced in ([5]) is valid only in the case when equation (6) holds.

Direct experimental confirmation of the importance of introducing the criterion $\mathrm{Re}_e$ follows from the work of Bagley ([4]), who calculated the magnitudes of elastic deformations at the onset of the critical flow regime on the basis of experiments carried out with three polymers (polyethylene, polystyrene, polymethyl methacrylate). The quantity $\gamma_k$ is equivalent to $\mathrm{Re}_{ek}$ for the case of flow in a capillary. Therefore, the experimentally discovered constancy of $\gamma_k$—using as examples polymers very different in their physical and chemical nature and at different temperatures—serves as confirmation that $\mathrm{Re}_e$ is indeed a criterion for the onset of elastic turbulence. The small deviations of $\gamma_k$ from the mean value, approximately equal to 7.0, when evaluated criterially, lie within the error limits of the experiments themselves in determining the onset of the critical flow regime.

Thus, numerous published experimental results well confirm the phenomenological theory developed in the present work for the onset of elastic turbulence. Analysis of only the first of the complete set of criteria has already made it possible to give the correct picture of the onset of spontaneous disturbances of shear-deformation regimes in elastic-viscous media. Taking the subsequent criteria into account may make it possible to refine the picture and allow for secondary phenomena.

The system of criteria $A_k$, $B_k$ and, in particular, $\mathrm{Re}_e$ can be used not only in the analysis of flow in a capillary, but also in other cases of deformation of elastic-viscous systems.

The authors express their gratitude to G. V. Vinogradov for supervising the work and to Acad. V. A. Kargin for discussing it.

*Note added in proof.* In a recently published work ([15]), it has been experimentally proved that, irrespective of the place where unstable flow originates (the entrance to the capillary or the surface of the capillary itself), the phenomenon under consideration nevertheless arises at a constant value of the accumulated elastic deformation $\gamma_k \approx 5$, which is comparable with Bagley' s results ([4]).

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Received
21 III 1963

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*Note: Figure translations are in progress. See original paper for figures.*