CONDITIONS FOR THE OCCURRENCE OF ELASTIC TURBULENCE IN POLYMER SOLUTION FLOWS THROUGH POROUS MEDIA

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The occurrence of elastic turbulence and accompanying resistance anomalies in filtrational polymer solution flows is investigated. The critical value of the Weissenberg number is determined for a porous medium composed of spheres of the same size.

One of the reasons for interest in the hydrodynamics of polymer solutions is the unusual behavior of solutions during flow through porous media. One of the effects observed here, namely, an abnormal rise in the filtrational resistance of polymer solutions as compared with the resistance of the solvent, has found wide application in enhancing of oil recovery [1, 2]. The most significant (up to two orders of magnitude) deviations of resistance from the Darcy law were observed for the flows of dilute solutions of polymers of high molecular weight whose viscosity is close to that of the solvent [3-8].

It has long been known that anomalies in the filtration of polymer liquids are associated with their elastic properties [4, 9]. However, investigators faced great difficulties when they tried to explain the mechanism responsible for the increase in the viscous dissipation in filtrational flows of polymer solutions [3, 10]. In the case of dilute solutions, investigations were also complicated by the absence of experimental techniques for determining the elastic characteristics of such liquids. Attempts were made to use for calculations elastic characteristics of the theory that are based on molecular models of dilute polymer solutions that assume the indivisibility of macromolecules in the solutions [7, 8, 11, 12]. However, it was shown experimentally that the critical values of dimensionless combinations of the Deborah number type introduced by means of these theories to describe the occurrence of filtration anomalies depend on the concentration of the solution. This fact is not surprising in light of data indicating a relation between the elastic properties of dilute solutions of high polymers and supermolecular structures typical of these solutions [13].

A considerable increase in understanding filtration anomalies was achieved upon investigating the flow of polymer solutions in simple models of pore channels, namely, channels with a periodically varying cross section [14, 15]. It was shown [16, 17] that growth in resistance during filtration of polymer solutions was caused by elastic turbulization of the flows. The criterion characterizing the occurrence of elastic turbulence was taken to be the Weissenberg number

We =
$$\frac{\Theta v}{d}$$
. (1)

Here Θ is the natural time of the liquid, i.e., the time of relaxation of elastic stresses with low frequencies of perturbations, and v and d are the characteristic velocity and dimension of the channel of variable cross section. For channels with a square cross section the critical value of the Weissenberg number was determined to be We^{*} = 4.7. In this case the hydraulic diameter of the narrow section of the channel was used as the characteristic dimension, and the mean velocity in this section was used as the characteristic velocity [18]. Based on the results obtained, a technique was developed for measuring the natural time of polymer solutions of low concentration. It

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Reference	E	D _p , cm	М	[ŋ]	t, °C	K	We*	A
[5]	0.389	$62.5 \cdot 10^{-4}$	$4 \cdot 10^{6}$		24	1.60	$2.6 \cdot 10^{-3}$	262
	0.372 0.377 0.368	0.011 0.022 0.045	9.4 · 10 ⁵	470	25	0.27	$3.9 \cdot 10^{-2} 3.2 \cdot 10^{-2} 4.5 \cdot 10^{-2}$	180
[3]	0.372 0.377 0.368	0.011 0.022 0.045	4.2 · 10 ⁶	1 <i>5</i> 00	25	1.60	$4.3 \cdot 10^{-2} 4.5 \cdot 10^{-2} 6.4 \cdot 10^{-2}$	180
	0.372 0.377	0.011 0.022	9.7 · 10 ⁵	2900	25	3.28	$3.3 \cdot 10^{-2} \\ 2.7 \cdot 10^{-2}$	180
[7]	-	0.05 0.1	-	2300	-	2.65	0.1 0.1	150
[8]	_	0.019 0.04	-	2100		2.41	$2.2 \cdot 10^{-2}$ $2.2 \cdot 10^{-2}$	211

TABLE 1. Critical Weissenberg Number for the Flow of PEO Solutions in Packings of Equally Sized Spheres

turned out that the natural time for the solutions of nonelectrolyte polymers, in particular, aqueous solutions of polyethylene oxide (PEO), changes proportionally to the square root of the mass concentration of the solution C:

$$\Theta = K \sqrt{C} . \tag{2}$$

Here K is a coefficient that depends on the temperature of the solution and the characteristic viscosity of the polymer, determined at low shear rates, $[\eta] = \lim_{C \to 0, \ \dot{\gamma} \to 0} [(\eta - \eta_s)/(c\eta_s)].$

The results obtained make it possible to pass to a discussion of polymer solution flows in more realistic models of natural oil containers than tubes of variable cross section – random packings of equally sized spheres as well as packings with spheres of variable size. For packings of equally sized spheres a large volume of factual material has been accumulated [3, 5, 7, 8]. As a rule, the molecular weight M or characteristic viscosity $[\eta]$ of the polymers used is given, as well as the mean size of the spheres D_p and the porosity of the packings E. Graphs yield the critical values of filtration velocity V_0 or interstitial velocity $V = V_0/E$ at which filtration anomalies occur. In the case of packings of equally sized spheres the Weissenberg number can be introduced naturally on the basis of the quantities V and D_p . One expects that in such packings, just as in tubes of variable cross section, elastic turbulence arises at a certain We number (constant for different concentrations). This is indicated, in particular, by the inversely proportional dependence of the critical value of V/D_p on the square root of the concentration observed in filtration of aqueous solutions of PEO [7, 8] (together with relation (2)). However, this conclusion cannot be drawn on the basis of available experimental data. The values of the critical Weissenberg number We* based on these data and listed in Table 1 differ significantly in works published by different authors.

There are two plausible explanations for this fact. First, in the works cited the polymers used are not always characterized adequately. Thus, when compiling Table 1, to estimate the natural time (the coefficient K in relation (2)) we used data of [18], where the values of K are given for aqueous solutions of PEO as functions of molecular weight and of characteristic viscosity at low shear rates. The values of $[\eta]$ were taken to be values presented in the

works discussed and probably not measured very accurately. In general, the determination of the characteristic viscosity is hardly a simple experimental problem. Equipment is required that would allow one to measure the viscosities of polymer solutions of extremely low concentration under conditions of nearly zero shear rates. Capillary viscosimeters, used, for example, in [3], do not comply with the indicated requirements; therefore, passage to the limit of zero shear rate and concentration in determining $[\eta]$ was done with the help of double geometric extrapolation of viscosities. Such a procedure could introduce an appreciable error into the value of $[\eta]$. Comparing the values of $[\eta]$ given in [3] with those presented in [19], which is specially devoted to the study of the dependences of viscosity on the shear rates of dilute solutions of PEO, we may conclude that the authors of [3] failed to determine accurately the characteristic viscosities of the polymers used.

The other and, evidently, the main source for the scatter of the values of We^{*} is the difference in the characteristics of the porous media themselves. Thus, in the works cited the authors assumed that they used random packings of equally sized spheres. Proceeding from data for filtration of Newtonian fluids, we may estimate the extent to which the sizes and shapes of the spherical particles were identical in [3, 5, 7, 8]. The resistance curves for filtration through packings of spheres are usually presented in Ergun dimensionless coordinates:

$$\operatorname{Re} = \frac{D_{\mathrm{p}} V}{\nu} \frac{E}{1 - E},\tag{3}$$

$$\lambda = \frac{D_{\rm p}}{L} \frac{\Delta P}{\rho V^2} \frac{E}{1 - E}.$$
⁽⁴⁾

Here Re is the Reynolds number, λ is the resistance coefficient, ν is the kinematic viscosity, L is the length over which the pressure drop ΔP is determined. At small Reynolds numbers the Darcy law holds, under which the law of the resistance of packings takes the form

$$\lambda \operatorname{Re} = \frac{D_{p}^{2} E^{3}}{(1 - E)^{2}} \frac{1}{k} = A.$$
 (5)

Here k is the permeability of the packing, A is a dimensionless constant that depends on the properties of the porous medium. It is known that for random packings of equally sized spheres the values of A lie within 150-180 [3, 20, 21]. These values were confirmed many times in experiments with rather large-sized spheres, when one could easily control their size and shape. Therefore, with a certain arbitrariness the variation of the quantity A within the indicated limits can be attributed to the random nature of the packing of spheres, and the porous media themselves can be considered similar.

It is necessary to determine more precisely the size that should be taken as the characteristic dimension of the porous medium when we investigate the occurrence of elastic turbulence. In works devoted to the flow of polymer solutions in channels of variable cross section it was shown that the appearance of resistance anomalies is determined by the dimensions of the narrow portions of the channels [15, 22]. We may assume that such a situation also pertains to real porous media, where elastic turbulence occurs in the narrowest pore passages. It is difficult to determine the precise size of these passages but, considering geometrically similar porous media, e.g., random packings of equally sized spheres, we may use D_p as the characteristic dimension in the Weissenberg number for purposes of simplification.

The last column of Table 1 lists the values of A for the packings of spheres used in experiments with PEO solutions. These values are estimates either from the given characteristics of the porous medium k, E, D_p or from the resistance curves. In some publications these values are such that it is impossible to consider the porous media used as packings of equally sized particles and thus to compare the critical Weissenberg numbers of the occurrence of elastic turbulence.

There is a distinct correlation between the values of We^{*} and A in Table 1. The more the value of A exceeds the values typical for random packings of equally sized spheres, the smaller the critical Weissenberg number. This

No. of the set	Number of spheres in the set	D _p , mm	Maximal deviation of size relative to D _p , %	Sieves used, mm
1	30	0.213	±16	0.20 0.25
2	56	0.300	±18	0.25 0.35
3	30	0.778	±4	0.75 0.85

TABLE 2. Sizes of Polystyrene Spheres According to Microscopy Data

seems to signify that with a decrease in the permeability of packings, pore passages of increasingly smaller dimensions appear in porous media, precisely in which elastic turbulence occurs.

To determine quantitatively the critical value of the Weissenberg number for the occurrence of filtration anomalies of polymer solutions in packings of equally sized spheres, an experiment was carried out. Moreover, an attempt was made to demonstrate the influence of nonuniformity in the size of the spheres on this effect.

Packings of polystyrene spheres encased in cylindrical containers with wire screens were used. The construction of the containers of the packings coincided with that used in [3]. The diameter of the packing container D and the packing length L were selected so as to ensure convenient measurement of liquid flow rate and pressure drop, but so that boundary effects could be eliminated. Liquid escaped from a pressure tank, the liquid level in which could be varied. The pressure tank and the conveying hose were fitted with thermostatic water jackets. This made it possible to conduct experiments at a constant temperature within $\pm 0.5^{\circ}$ C. Using a set of graduated cylinders and a stopwatch, we measured the volumetric liquid flow rate Q. The pressure drop ΔP over the length L was measured with the help of piezometers.

Special attention was paid to the preparation of polystyrene spheres. The requisite number of spheres were passed several times through two standard sieves whose hole size limited the size of the spheres from above and from below. Three sets of spheres were obtained that had a certain size which was controlled with the help of a measuring microscope by using samples of 30 or more spheres. Table 2 presents parameters of the homogeneous sets of spheres obtained. It is seen that sieving made it possible to obtain sets with a narrow size distribution.

In all, three packings were prepared. Two of these, containing spheres with $D_p = 0.300$ and 0.778 mm, were used for determining We^{*} in polymer solution flows in porous media composed of equally sized spheres. The third packing contained a mixture of large and small spheres and was used for investigating the effect of size nonuniformity. The small spheres were selected of such a size that they could fill the spaces between the large spheres. Assuming for simplicity a tetrahedral packing, the diameter of a small sphere that can be inscribed in the cavity formed by four large spheres can be easily estimated to be 22.5% of the diameter of the latter. Correspondingly, when the diameter of a large sphere is $D_p = 0.778$ mm, a small sphere should have a size of 0.180 mm. Unfortunately, the available set of sieves only permitted us to obtain spheres of a somewhat larger size with $D_p = 0.213$ mm, which, however, is of no importance within the framework of a qualitative experiment. The ratio of the mumbers of spheres was selected in such a way that small spheres could fill all the spaces with excess. The numbers of small and large spheres were in the ratio of 5:1, with the large ones occupying 90% of the overall volume of the spheres in the packing.

The containers of packings were filled with spheres with the help of vibration. The porosity E was calculated from the volume of the liquid that could fill the packings. In packings of equally sized spheres E = 0.37, which agrees with data of [3]. The packing with spheres of nonuniform size had a somewhat smaller porosity of E = 0.34.

After determination of *E*, experiments with Newtonian liquid flow through packings were conducted. Using the measured values of ΔP and the filtration velocity $V_0 = 4Q/\pi D^2$ we determined the permeability, which was



Fig. 1. Resistance to Newtonian fluid flow through a packing of equally sized spheres: $D_p = 0.778$ (1) and 0.300 mm (2).

Fig. 2. Dependence of the relative pressure drop on the interstitial velocity for the flow of aqueous solutions of PEO, $[\eta] = 2300$: $C = 5 \cdot 10^{-5}$ (1), 10^{-4} (2), $2 \cdot 10^{-4}$ (3), and $4 \cdot 10^{-4}$ (4), in a packing with $D_p = 0.300$ mm. V, cm/sec.



Fig. 3. Critical Weissenberg number of the occurrence of elastic turbulence for the flow of aqueous solutions of PEO, $[\eta] = 2300$, as a function of the concentration in packings of equally sized spheres: $D_p = 0.778$ (1) and 0.300 mm (2).

Fig. 4. Dependence of the relative pressure drop on the interstitial velocity for a PEO solution flow, $[\eta] = 2300$, $C = 3 \cdot 10^{-4}$, in packings with $D_p = 0.778$ (1) and 0.300 (2) and in a packing with a mixture of spheres of two sizes (3).

equal to $7.03 \cdot 10^{-7}$, $4.80 \cdot 10^{-6}$, and $1.91 \cdot 10^{-6}$ cm² for the packings with $D_p = 0.300$ and 0.778 mm and the packing with a mixture of spheres, respectively. The values of Re and λ were calculated from Eqs. (3) and (4). The resistance curves for the packings of equally sized spheres are given in Fig. 1; the points cluster about the single curve $\lambda = A/Re$, where A = 164. We may assume that the packings used are really random packings of equally sized spheres.

In the experiments we used solutions of PEO in distilled water. The value of $[\eta]$ determined with the aid of a Zimma-Krothers viscosimeter was equal to 2300. The solutions were prepared identically as described in detail in [19]. With the help of channels of variable cross section [16], the natural time of solutions was measured at a working temperature of $t = 22^{\circ}$ C. To prevent the degradation of the polymer solutions, measurements were made with a single passage of the liquid though the packings.

To determine the critical value of the interstitial velocity V^* at which elastic turbulence occurs, it is convenient to present the results in the form of the dependence of the relative pressure drop $\Delta P/\Delta P_0$ on V. Here ΔP_0 is the pressure drop in the flow of the solvent calculated from the Darcy law. Figure 2 presents relationships obtained for the flow of PEO solutions, $[\eta] = 2300$, of different concentrations through a packing with $D_p = 0.300$ mm. Upon the occurrence of elastic turbulence we observed a rather sharp deviation of the value of $\Delta P/\Delta P_0$ from the horizontal line corresponding to laminar flow. Similar relationships were also observed for a packing with $D_p =$ 0.778 mm. Using a certain value of V^* , the critical values of the Weissenberg number were calculated as functions of the mass concentration of the solutions (Fig. 3). The points cluster about the mean value We^{*} = 0.16. With experimental accuracy the value of We^{*} is a constant independent of the size of the spheres and the properties of the solutions. Since the porous media and the viscoelastic fluids used are rather fully characterized in the present work, we may regard the problem of determining the critical parameter responsible for the inception of elastic turbulence in random packings of equally sized spheres to be solved.

To investigate the flow in a packing containing spheres of two sizes, we prepared a sufficient quantity of a PEO solution with a concentration of $C = 3 \cdot 10^{-4}$ and determined its natural time Θ . We determined the critical values of the interstitial velocity for the flow of this solution through all three packings (Fig. 4). It turned out that the influence of the addition of small spheres is very great. Elastic turbulence in a packing with spheres of nonuniform size occurs at interstitial velocities close to the values for a medium with $D_p = 0.300$ mm. It may seem that there are no large spheres in such a packing even though they occupy almost the entire volume.

The following qualitative explanation of this fact is possible. It was already noted that elastic turbulence of flows occurs first of all in the narrowest pore passages, where the maximum rates of deformation of the liquid are observed. When small spheres are added to a porous medium made up of large spheres, still smaller passages are formed in it. As a result, the deformation rate corresponding to the occurrence of elastic turbulence is attained in the narrow passages of a medium with a spreed of sizes at smaller values of V than in a medium with identical large spheres.

NOTATION

 Θ , natural time; v, characteristic velocity; d, characteristic size; We, Weissenberg number; C, mass concentration; K, coefficient in the empirical relation for Θ ; $[\eta]$, characteristic viscosity based on the first Newtonian viscosity; D_p mean size of spheres; E, porosity; V_0 , filtration velocity; V, interstitial velocity; *, critical value of a parameter; Re, Reynolds number; A, constant in the resistance law; λ , resistance coefficient; L, packing length; ΔP , pressure drop; v, kinematic viscosity; k, permeability; Q, volumetric liquid flow rate; D, packing diameter.

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