STRUCTURE OF WEAK SOLUTIONS OF POLYMERS SHOWING THE EFFECT OF TURBULENCE DAMPING

G. N. Barenblatt, I. G. Bulina, V. N. Kalashnikov, and N. M. Kalinichenko

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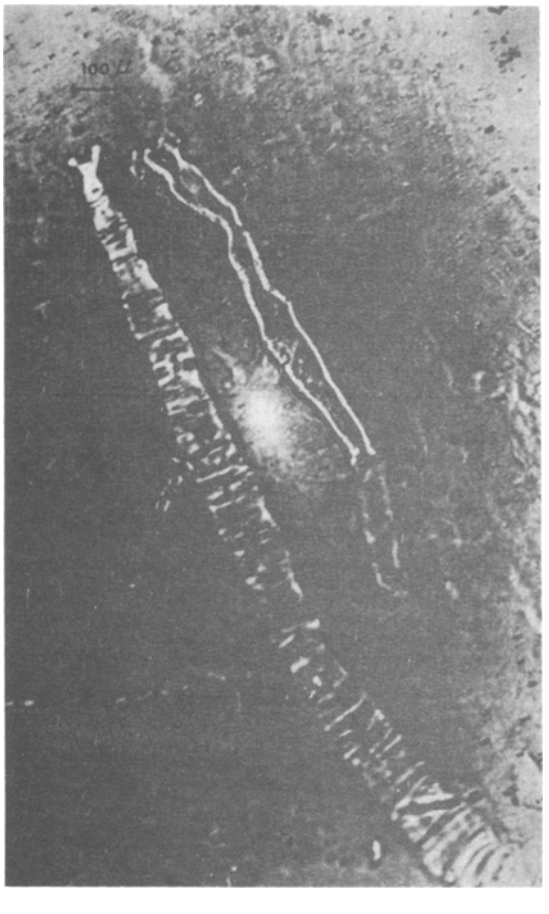
An appreciable reduction in resistance to turbulence was found in [1–9] when very small amounts of certain polymers were added to the fluid. Studies of dyes injected into such flows have shown [10, 11] that, at least in some cases, the drag reduction is associated with turbulence damping. Since the size of the polymer molecules is negligible in comparison with the size of the smallest eddies in the flow (the Kolmogorov internal scale of turbulence), one of the possible explanations of the effect [12] is that the polymer molecules capture the solvent molecules, producing relatively large and rigid hypermolecular formations with linear dimensions comparable with the internal turbulent scale, i.e., capable of affecting the turbulence. This hypothesis was indirectly confirmed by the rapid fall in the permeability of filters and capillaries for aqueous solutions of a sodium salt of carboxymethylcellulose when the diameter of the filters and capillaries was reduced down to a figure of the order of the Kolmogorov scale in the flows investigated in [8, 10]. Control experiments using a Newtonian liquid of the same viscosity (aqueous solution of glycerin) did not show this anomaly. In this note we report the results of microscopic observations on water solutions of the sodium salt of carboxymethylcellulose (CMC), which confirm directly the presence of hypermolecular formations.

The solutions were prepared by dissolving dry CMC in water at room temperature. A polarizing microscope was used for the microscopic studies. Observations were carried out in polarized and ordinary light at a magnification of 50–150. The solution was placed under the microscope on a glass slide, or in a glass capillary, and the observations were carried out both on a stationary solution and on a solution flowing along the capillary. Specially prepared circular (diameter up to 1.5 mm) and square cross-section capillaries (side lengths 0.8, 0.5, and 0.2 mm) were employed. The flat-wall capillaries were formed from two flat plates separated by foil strips of the necessary thickness. The motion of the solutions was investigated at a velocity of 1 cm/sec. It was photographed at a rate of 100 frames/sec.

When the CMC solutions were observed under the microscope, it was found that they contained transparent formations with a refractive index equal to that of water. This meant that they could be seen even without special visualization procedures. However, still better visibility could be achieved by means of a suitable dye. When the dye was introduced, the new formations showed a darker color than the ambient liquid. However, the best results were obtained with polarized light, because the formations exhibited the phenomenon of birefringence. Figure 1 shows a photograph of the most characteristic formations obtained in polarized light. It was found that the lateral size of the formations was 50–200 μ, whereas the longitudinal size ranged up to 1000 μ in some cases. The formations were clearly periodic–there was an alternation of bright and dark bands. All this enabled us to designate this structure in accordance with established terminology as fibrillar (the term "globule" used in our previous paper [12] was, in this sense, inappropriate for hypermolecular structures in the CMC solutions).

The photographs show spindle-shaped fibrils with the characteristic two loops at the ends, and also short fibrils of differing length and with an appearance suggesting that they are bits of longer fibrils rather than complete structures. A characteristic longitudinal band runs along the axis of the fibrils. This suggests that the fibrils have an internal cavity, which was confirmed by direct observation under a microscope by viewing pieces of the fibrils at different angles. These pieces are short tubes whose internal diameter is smaller by a factor of about two than the external diameter. The structure of the fibril walls is clearly seen in polarized light between crossed polarizers, and when the angle between the fibril axis and the plane of polarization corresponds to the maximum darkening of the fibril. The fibril walls are then seen to consist of spirals packed in parallel. Usually there are two such spirals, sometimes three. When the solvent is evaporated from a drop of the CMC solution placed under a microscope, it is found that the fibril spirals contract, the diameter of the fibrils decreases substantially and their length contracts somewhat until the addition of water restores the previous dimensions. This shows that aqueous solutions of CMC are, in fact, colloidal solutions whose particles are highly saturated by the solvent. When the solution flows, the fibrils orient themselves so that their longitudinal axes lie along the flow lines. The observed reduction in the CMC viscosity with increasing flow rate [13] can probably be explained by the alignment of the fibrils in the flow.



The fibrils are elastic particles and can be substantially deformed without fracture by external forces. They return to their original state when the load is removed. On one of the motion-picture records we saw a case in which a fibril approached the mouth of the capillary laterally and covered the entrance. As a result of the pressure drop it then bent into two, and in this form entered the capillary. However, at the exit of the capillary it straightened out again, assuming the original straight-line shape.

The fibrils can also be elastically deformed in the axial direction. A situation was recorded where the cross section of an 0.2-mm diameter capillary was covered by a coaxial fibril. The pressure difference which appeared in this situation led to a small contraction of the length of the fibril. Here again, the fibril returned to its initial state when the pressure was removed. The elasticity of the fibrils may be the reason for the observed lateral elasticity of CMC solutions [14].

As noted above, the fibrils are transparent forms showing birefringence. Their optical axis lies along the major symmetry axis. The

orientation of the fibrils in the flow should lead to a special optical direction for the flow as a whole, i.e., the flow should show the integral birefringent effect. Flows of many polymer solutions do, in fact, exhibit birefringence [15], and it would appear that one of the reasons for this is the presence in the solution of anisometric birefringent formations which are aligned during flow. The fibrils contain a relatively large amount of water, which increases with decreasing concentration of the solution. The fibril diameter increases with decreasing concentration, while the fibril brightness under the microscope between crossed polarizers decreases. At concentrations of the order of 1% by weight, the amount of water in the fibrils is considerably less than in the case of high dilution, but remains quite high, which is indicated by the substantial reduction (by a factor of 4-5) in the diameter and a certain contraction of the fibril length on drying.

The considerable immobilization of a given part of the solvent by a polymer may explain the fact that very small additions of certain polymers have a very considerable effect on the hydrodynamic parameters of the solutions. A small polymer impurity leads to the appearance of rigid and large formations in the solution, consisting largely of the solvent molecules. These are sufficient in quantity to produce a hydrodynamic effect on the flow and, in particular, to reduce the intensity of fluctuations in turbulent flows.

There may be several possible mechanisms responsible for the additional turbulence damping due to the hypermolecular formations which we have established.

Firstly, the damping may be connected directly with the relative motion of fibrils in the surrounding fluid, and their eleastic deformation by small eddies, followed by the conversion into heat of the elastic vibrations of the fibrils. In an unpublished report M. Tulin (Hydronautics Inc., USA) has suggested the following very interesting hypothesis about the turbulence damping mechanism. According to this hypothesis turbulence damping is connected with the fact that under the action of high shear rates the long molecular spirals in the flow straighten out, take up elastic energy, and then release this energy into transverse waves whose velocity of propagation is comparable with fluctuations in the flow velocity. In our opinion, this phenomenon may occur not at the molecular level but at the hypermolecular level, so that the globules or fibrils are deformed rather than the individual polymer molecules.

Secondly, the globules or fibrils form a surface film on the walls of the tube or the body past which they flow, and can thus affect the formation of eddies in the boundary region. They also substantially damp the eddies in this region due to relative fibril oscillatory motions or deformations, and the formation of shear waves.

The precise mechanism through which small polymer impurities affect fluid flows demands, of course, additional study, but it may be regarded as established that hypermolecular structures must be taken into account in the study of turbulent flows of weak polymer solutions.

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Scientific Research Institute of Mechanics, Moscow State University

Institute of Problems in Mechanics, AS USSR