Spontaneous Growth of Fluctuations in the Viscous Flow of a Fluid past a Soft Interface

V. Kumaran and R. Muralikrishnan
Department of Chemical Engineering, Indian Institute of Science, Bangalore 560 012, India
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The flow induced instability in the flow past a soft material is studied in the limit of low Reynolds number where inertial effects are insignificant. A transition from laminar flow to a more complicated flow profile is observed when the strain rate of the base flow increases beyond a critical value; the transition is found to be reproducible. The experimental results are compared with theoretical predictions and quantitative agreement is found with no adjustable parameters.

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Fluid flow in biological systems at micron scales is characterized by two salient features. The first is that the Reynolds number $\rho VL/\eta$, which is the ratio of inertial to viscous effects, is typically small. Here, ρ and η are the fluid density and viscosity, V is the characteristic velocity, and L is a characteristic length scale. For example, the flow of blood of viscosity is 10^{-2} kg m^{-1} s^{-1} in microcapillaries of diameter 10 μm at typical velocities of 1 cm s^{-1}, the Reynolds number is 0.01, and at smaller scales and lower velocities the Reynolds number could be even lower. This implies that fluid inertia is not a factor in determining the flow dynamics, in contrast to typical industrial applications where inertia has a dominant effect. The second salient feature is that the flow occurs past surfaces which are soft and flexible, and the elasticity of the surfaces could affect the flow. Tissue in biological systems have a coefficient of elasticity of about $10-10^2$ kg m^{-1} s^{-2}, which is about 3 to 4 orders of magnitude lower than that of surfaces typically encountered in industrial applications. Consequently, biological flows could be influenced by the balance between the viscous stresses due to fluid flow and the elastic stresses due to the deformation of the surfaces bounding the flow. The ratio of these is given by the dimensionless parameter $(V\eta/GR)$, where G is the modulus of elasticity of the wall material. For typical parameter values listed above, this dimensionless variable could be $O(1)$.

An important feature of flow in channels and pipes is the transition from laminar to turbulent flow. This is important in practical situations because the drag force and transport coefficients for a turbulent flow are higher than those for a laminar flow with the same mean velocity. The transition in the flow through rigid tubes and channels has been extensively studied, and it is well known that the transition occurs when the Reynolds number exceeds a critical value of $O(10^3)$ (the exact value depends on the flow geometry). The transition from a laminar to a more complex flow could also be important in biological systems, since this could have a significant influence on the transport of oxygen, nutrients, and waste matter. As mentioned above, inertial effects are negligible at micron scales in biological systems, but a balance between viscous and elastic stresses could destabilize the laminar state of the flow. There have been numerous studies on the stability of fluid flow past compliant surfaces [1–3]. However, most of these studies have considered high Reynolds number flows where inertial effects are significant, since they have been motivated by marine and aerospace applications. There are a few applications, such as hollow fiber reactors and membrane bioreactors, where flow past soft materials are encountered. Krindel and Silberberg [4] conducted experiments on the flow of a fluid through a tube whose walls were made of polyacrylamide gel. Though they reported that the elasticity of the wall appeared to reduce the Reynolds number at which the transition takes place, these experiments were carried out at Reynolds number of $O(100)$ where inertial effects are significant.

The stability of the laminar flow of a Newtonian fluid past a flexible surface in the absence of inertia was studied in different geometries by Kumaran *et al.* [5] and Kumaran [6]. These studies predicted that the laminar state of the flow does become unstable when the parameter $(V\eta/GL)$ exceeds a critical value. The present experiments provide the first experimental verification of the transition in the absence of inertia.

The fluid used in the experiments was commercial grade silicone oil manufactured by GE silicones. The polyacrylamide gel, which was used for the flexible surface, was prepared as follows. The typical concentrations of the constituents of the gelation mixture were N-acrylamide (monomer) 5%, bis-acrylamide (cross-linker) 0.05%, ammonium persulfate (initiator) 0.12%, and TEMED (catalyst) 0.062% in water. The concentrations of the catalyst and initiator were kept constant throughout the experiments, while the concentrations of the monomer and cross-linker were changed in the range 5%–8% and 0.05%–0.08% to modify the viscoelastic properties of the gel. The gel was cast in the form of a rectangular sheet of thickness 4.5 mm and side 5 cm × 5 cm between two glass plates. No surface modification was made at the gel surface. In order to compare the results of the experiments with theoretical predictions, it is necessary to determine the storage modulus G' and the loss modulus G'' as a function of frequency. These were determined using a parallel plate geometry in the Rheolyst AR1000 rheometer

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described below. It was found that the storage modulus G' shows a distinct plateau region in the frequency range 0.1 to 10 Hz. The plateau value of the storage modulus was used for comparison with theory. Experiments indicated that in the frequency range between 1 to 10 Hz, the loss moduli of the gels showed a scaling law $G'' \propto \omega^{1/2}$, and this form of the loss modulus was used for comparison with theoretical results.

The experiments were carried out using a Rheolyst AR1000 rheometer using a parallel plate geometry. The geometry consists of a stationary lower plate which has temperature control and normal force measurement capabilities, and a rotating upper plate which is disk shaped and has a radius of 2 cm, as shown in Fig. 1. The procedure used for measuring the viscosity of a fluid is as follows. (1) The fluid is placed on the lower plate, and the upper plate is lowered until a preset distance between the two plates (300–1000 μm) is achieved. The fluid forms a thin film between the two plates which is held in place by surface tension. (2) The upper plate is then rotated, and the angular velocity and torque are measured. (3) The stress and strain rate at the outer edge are then calculated by the rheometer software, *assuming the flow is laminar,* and the ratio of these provides the fluid viscosity. It is important to emphasize that the viscosity of the fluid is calculated *assuming the flow is laminar,* and an anomalous increase in the apparent viscosity reported by the rheometer is an indicator of a transition to a more complex flow pattern.

For the present experiments, this geometry was modified as shown in Fig. 1(b). (1) Polyacrylamide gel was cast as described above, and this sheet was placed on the lower plate. (2) The upper plate was lowered until it made contact with the polymer gel, and the condition for contact was considered to be a normal force of 0.2 N; this value of the normal force was chosen because it is the same value used for determining contact with the stationary lower plate in viscosity measurement experiments. The micrometer reading at contact provided the thickness of the gel. (3) The upper plate was then raised, and the fluid was placed on top of the gel. (4) The upper plate was lowered until a gap of the desired thickness (between 300 μm and 1 mm for the present experiments) of the fluid film between the upper plate and the gel surface was attained. (5) The rheometer was operated in the stress controlled mode, where the torque on the top plate was increased linearly with time, and the apparent viscosity (which is the ratio of the stress and strain assuming that the flow is laminar) was recorded.

The results of a typical experiment are shown in Fig. 2, where the *apparent viscosity determined assuming the flow is laminar* is plotted as a function of strain rate. The temperature of the plates was set equal to 28 °C, and the temperature was monitored throughout the experiment to ensure that there was no viscous heating. The rheometer was operated in the stress controlled mode, where the torque on the top plate is increased at a constant rate and the angular velocity is recorded. The shear stress and strain rate at the outer edge are calculated assuming the flow is laminar. In this experiment, the rate of increase of torque corresponded to a rate of increase of 4 Pa/s of the stress at the outer edge. The broken line in Fig. 2 shows the apparent viscosity for the flow between two rigid surfaces as shown in Fig. 1(a). It is seen that there is a small decrease in the apparent viscosity at low shear rates, probably due to instrument limitations in the measurements of low torques. After this, the viscosity settles to a constant value up to a strain rate of about 1500 s^{-1}. The solid line shows the strain rate and viscosity for a different experiment performed in the configuration Fig. 1(b) with the same thickness of the fluid layer and rate of increase of shear stress. It is observed that the apparent viscosity is close to that of the fluid when the strain rate is less than about 750 s^{-1}, but there is a sharp increase in the apparent viscosity at this point and simultaneously there is a decrease in the strain rate. This indicates that the flow in the gap is no longer

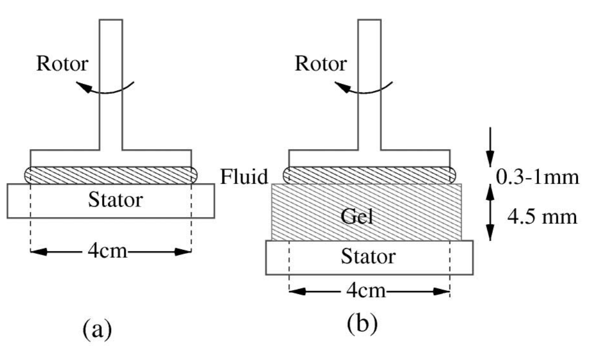

FIG. 1. Rheometer used in the experiments. (a) Unmodified; (b) modified.

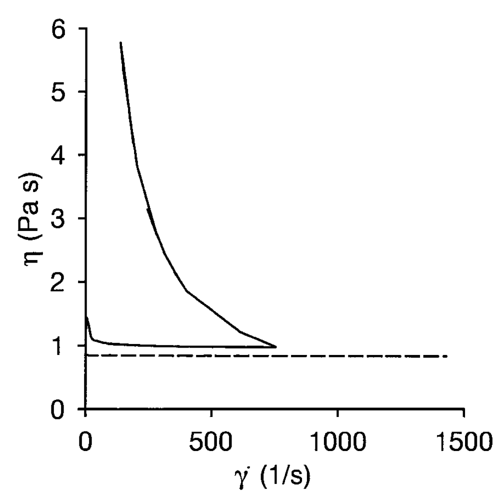

FIG. 2. Apparent viscosity η (Pa s) as a function of strain rate $\dot{\gamma}$ (1/s) for the flow past a gel of shear modulus 2354 Pa and thickness 4490 μm. The fluid thickness is 300 μm, and the shear rate was increased at the rate of 4 Pa/s. The broken curve shows the result for the same fluid thickness between rigid surfaces.

laminar, but a transition has occurred to a more complicated flow profile due to an instability in the laminar flow. The decrease in the strain rate accompanying the increase in viscosity is due to the fact that the rheometer is operated in a stress controlled mode, where the product of the viscosity and strain rate is constrained to increase at a constant rate. However, it should be noted that after transition, the apparent viscosity and strain rate measured by the rheometer are not indicative of the microscopic values in the fluid, because the flow is no longer laminar. Therefore, only the value of the strain rate in the laminar flow at the point of transition has physical significance, and this will be compared with theoretical results. After transition, the shear stress due to this flow increases to a sufficient extent that it exceeds the yield stress of the gel. At this point, small bits of the surface of the gel break off, and are suspended in the liquid film, and the gel surface becomes irregular.

Though the instability in the flow causes irreversible damage to the gel, the onset of instability can be measured reproducibly if care is taken to ensure that the experiment is stopped before damage occurs. This is shown in Fig. 3, where the onset of instability is shown for three different runs on the same gel. The strain rate for the onset of instability (where there is a sharp increase in viscosity accompanied by a decrease in the strain rate) is found to be unique for a given gel and gap thickness, and is independent of the rate at which the stress is increased. This is observed in Fig. 3, where the strain rate for transition remains unchanged while the rate of increase of stress is varied from 8 to 0.25 Pa/s. This suggests that the instability is initiated by the interaction between the fluid flow and the gel dynamics. In addition, the strain rate for the onset of instability is independent of the rate at which the stress is increased, as shown in Fig. 3.

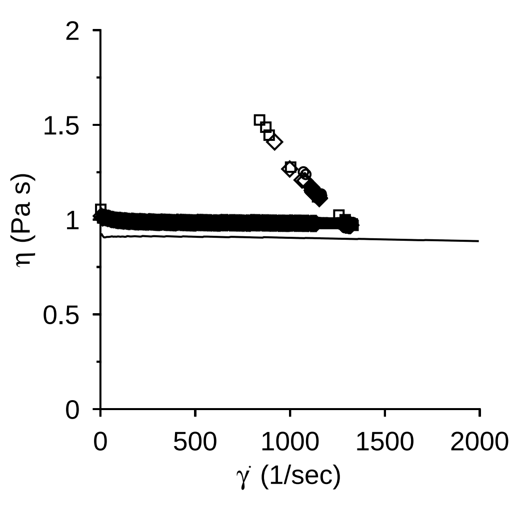

FIG. 3. The variation of the apparent viscosity η with the strain rate in a controlled stress experiment. The solid curve shows the viscosity as a function of strain rate for the flow between two rigid surfaces. The symbols show the results for the flow past a gel of thickness 4600 μm and storage modulus 3113 Pa, and fluid thickness 500 μm. ∘: Shear stress increased at 8 Pa/s; □: Shear stress increased at 7 Pa/s up to 1250 Pa, and then at 0.5 Pa/s. ◇: Shear stress increased at 7 Pa/s up to 1250 Pa, and then at 0.25 Pa/s.

It is useful to determine the characteristic Reynolds number in this case, and verify that this is indeed small. The viscosity of the silicone oil is close to 1 $\mathrm{kg\,m^{-1}\,s^{-1}}$, and the density is close to that of water, 10^3 $\mathrm{kg/m^3}$. The maximum width of the gap was 1 mm in the experiments, and the maximum velocity of the outer edge of the plate was of the order of 10 cm/s. For these parameter values, the Reynolds number is, at maximum, 0.1. Since the geometry involves a rotating plate, it is also necessary to examine the ratio of centrifugal and viscous forces. The pressure due to centrifugal effects scales as $\rho\Omega^2R^2$ or ρV^2, where Ω is the angular velocity, R is the radius of the plate, and V is the velocity of the outer edge. The viscous stresses scale as $V\eta/L$, where L is the gap thickness. It can easily be verified that the ratio of these is equal to the Reynolds number for the present case, and therefore centrifugal effects are also small compared to viscous effects. The dominant factors affecting the stability of the flow are the viscous stresses in the fluid and the elastic stresses in the gel. Moreover, the response time for the gel scales as (η_g/G'), which is $O(10^{-2}\text{ s})$ for the present case, indicating that transients decay over time scales small compared to the time scale for increase of the stress.

The experimental results are compared with the theoretical studies [5] of the two dimensional Couette flow between a moving rigid wall and a stationary viscoelastic surface. It is appropriate to assume the flow is two dimensional near the outer edge because the diameter (4 cm) of the top plate is large compared to the gap thickness (0.3–1 mm). The nondimensional parameters in the theoretical study are the ratio of the shear stress and shear modulus $(\tau/G') = (V\eta/G'R)$, the ratio of thicknesses (H/R), and the ratio of viscosities (η_g/η). Here, G' and $\eta_g = (G''/\omega)$ are the storage modulus and viscosity of the gel, H and R are the gel and fluid thickness, η is the fluid viscosity, and V is the maximum velocity at the outer surface of the top plate. The storage and loss moduli for the gel were obtained using the rheometer explained above, and the viscosity of the fluid η was determined using the rheometer. The thickness of the gel H was determined using the normal force facility in the rheometer, and the gap thickness R was preset to a desired value. Therefore, a comparison can be made with no adjustable parameters.

The theoretical analysis [5] was a linear stability analysis of the flow of a Newtonian fluid past a polymer gel. The configuration consists of a film of fluid bounded by a stationary polymer gel, and a flat plate moved at a constant velocity. In the base state, a linear velocity profile was set up in the fluid, and the stability of this flow to perturbations was analyzed using a linear stability analysis. The analysis predicts that perturbations become unstable when the strain rate in the base flow exceeds a critical value, which depends on the parameters listed above. The mechanism of

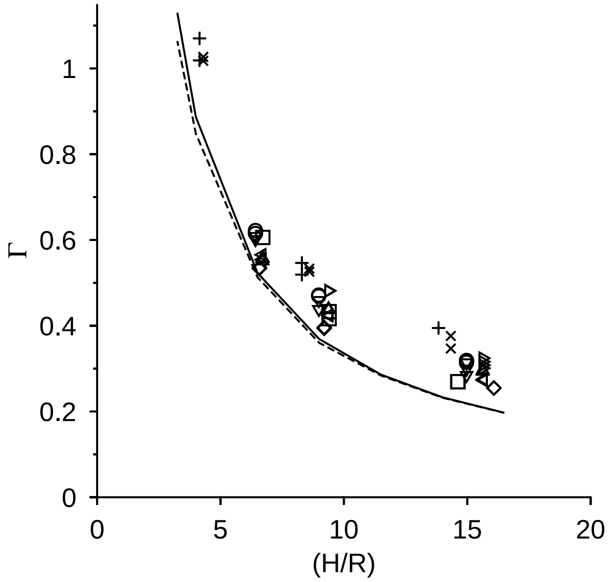

FIG. 4. The ratio (τ/G') of the transition value of the shear stress τ and the shear modulus G' as a function of the ratio of fluid and gel thicknesses (H/R) for different gels. Solid line: theoretical prediction for $G' = 4000$ Pa; broken line: theoretical prediction for $G' = 1000$ Pa; $\circ$: $H = 4490\ \mu$m, $G' = 2305$ Pa; $\square$: $H = 4699\ \mu$m, $G' = 3788$ Pa; $\diamond$: $H = 4600\ \mu$m, $G' = 4214$ Pa; $\triangle$: $H = 4690\ \mu$m, $G' = 2642$ Pa; ∇: $H = 4490\ \mu$m, $G' = 2354$ Pa; $\triangleleft$: $H = 4678\ \mu$m, $G' = 4040$ Pa; $\triangleright$: $H = 4690\ \mu$m, $G' = 3595$ Pa; $+$: $H = 4150\ \mu$m, $G' = 947$ Pa; $\times$: $H = 4300\ \mu$m, $G' = 1027$ Pa.

instability is the transfer of energy from the mean flow to the perturbations due to the shear work done by the mean flow at the surface. According to the linear stability analysis, the ratio (τ/G') for the onset of instability is a constant for a given ratio of thicknesses (H/R). This ratio, which is shown as a function of (H/R) in Fig. 4, is found to vary by about 5%–7% for a large number of gels, even though the shear modulus varies by a factor of 4. Figure 4 also shows a comparison between the experimental results and the theoretical predictions [5], and excellent agreement is found with no adjustable parameters.

The present study provides experimental evidence of the onset of a flow instability due to the fluid flow past a soft material. The experimental results are in close agreement with theoretical predictions, confirming that the instability is induced by the transport of energy from the mean flow to the fluctuations due to the shear work done at the surface [5,6]. The important result of the present study is that the effect of fluid flow past a soft material is different from the effect of an equal shear stress applied by other means. The coupling between the flow dynamics and the deformation of the material results in the spontaneous growth of perturbations. This could have important implications in the study of the effect of fluid flow in biological systems.

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