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Abstract

The present paper is a thorough study of the flow regime where an asymmetry of the mean axial velocity profiles is observed for shear-thinning fluids flow in a pipe [**?**]. This study is based on statistical analysis of the axial velocity fluctuations. It is shown that this flow regime exhibits features of a weak turbulence: chaotic in time and regular in space. More precisely: (i) power spectra of axial velocity fluctuations decay following a power law with an exponent very close to -3, (ii) large-scale coherent structures are generated and (iii) there is essentially no intermittency in this flow regime.

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Introduction. Transition to turbulence for Newtonian fluid in a pipe flow is still an important fundamental and practical problem since Reynolds's (1883) original experiments. It is ruled by the nonlinear inertia term in the equation of motion. Numerous experimental and numerical studies were carried out after this pioneering work. A recent literature review can be found in [?]. From mathematical point of view, pipe flow is linearly stable, yet in practice, above a critical Reynolds number an abrupt transition to turbulence occurs: Turbulent localized patches (so-called turbulent puffs [?]) generated by the sharp entry geometry move along the tube at approximately the bulk flow speed. The details on the structure of the turbulent-puff have been recently revealed [?]. The azimuthal features of a cross-sectional view are very similar to the travelling waves discovered by Faisst and Eckhardt [?] and Wedin and Kerswell [?]. These new solutions, based on the self-sustaining process proposed by Waleffe[?] and the subsequent continuation approach [?], are thought to connect to form an attractor.

Concerning the transition to turbulence for non-newtonian fluids, very little is available in the literature, despite the importance of this problem in the design and control of several industrial processes, such as in oil-well cementing, extrusion of molten polymers, paper coating, etc. Nevertheless, the existing literature reveals an interesting and yet unexplained effect. In a certain range of Reynolds numbers, the mean flow presents an asymmetry, while in the laminar and turbulent regimes the flow is axisymmetric. Here, mean refers to time-averaged. The departure from axisymmetry was observed independently by Escudier and Presti [?] for a flow of Laponite suspension (thixotropic shear-thinning fluid) at $1275 \leq Re_w \leq 3000$ and Peixinho *et al.* [?] for a flow of Carbopol (shear-thinning yield stress fluid) at $1800 \leq Re_w \leq 4000$. The Reynolds number Re_w is defined with the wall shear viscosity, the diameter D of the pipe and the bulk velocity $\hat{W}_b$. These two groups jointly published these and additional observations in [?]. The results of such asymmetric velocity profiles in the transitional regime are reported for a wide range of shear-thinning polymer solutions: Xanthan gum, polyacrylamide, Carboxymethylcellulose, and also for Carbopol and Laponite. For a Newtonian fluid (aqueous Glycerine solution) the time averaged velocity profiles measured are invariably axisymmetric. Further observations of asymmetry in the transitional pipe flow of Xanthan gum and Carbopol solutions have been reported by Guzel *et al* [?]. It was clearly indicated in [?] that this asymmetry must be a consequence of a fluid-dynamics mechanism rather than imperfections in the flow facilities. This mechanism has to be linked to the non-newtonian behaviour of the fluid. All non-newtonian liquids investigated display two common rheological properties: shear-thinning and viscoelasticity. Actually, the experimental results indicate that the asymmetry is shear-thinning dependent. Indeed, non-newtonian liquids with similar shear-thinning and different elastic behavior show similar degree of asymmetry, while elasticity seems not to influence the asymmetry [?]. In addition, the degree of asymmetry increases with increasing the shear-thinning. These conclusions are supported by a direct numerical simulation of pipe flow of shear-thinning fluids (power law model), at moderate Reynolds numbers ($5000 < Re_w < 8000$), performed by Rudman *et al.* [?]. The authors indicated that "the active region of the flow continually moves along the pipe to preferentially occur at one azimuthal position for extended times". We think that these computations were done for Re_w where there is an intermittency between the nonlinear asymmetric state and the fully turbulent regime, cf. Fig. 6 in [?]. The azimuthal structure of the asymmetry together with its axial position were first investigated by Esmael and Nouar [?]. The experimental results obtained suggest the existence of a robust nonlinear coherent structure characterized by two weakly modulated counter-rotating longitudinal vortices.

The previous articles focused mainly on the description of the asymmetry of the mean axial velocity profiles observed for different shear-thinning fluids. The purpose of the present study is to provide a deeper analysis of the flow regime where the asymmetry is observed, from a complete statistical analysis of velocity fluctuations. It is shown that this nonlinear asymmetric state is a weakly turbulent flow, i.e. chaotic in time and regular in space [?] driven by the nonlinear variation of the effective viscosity μ with the shear rate $\dot{\gamma}$, bringing into play the interplay between inertia and shear-thinning. Before presenting the experimental results one has to note that the Hagen-Poiseuille flow of yield-stress shear-thinning fluids is linearly stable for all Reynolds number and the optimal perturbations of the transient growth theory at the same dynamical and rheological parameters as in the experimental tests bear no resemblance to experimental observations [?].

Experimental set-up, instrumentation and tested fluid. Full details of the flow facility and instrumentation have been given in [?] and so only a brief description is provided here. The measurements were carried out in a plexiglass tube 30 mm inner diameter and 4.5 m long from the inlet (150 diameters). The velocity measurements were made using a Dantec FlowLite LDA system with a measuring volume 0.65 mm in length and 77 μm in diameter.

The fluid used is a 0.2w% neutralized aqueous solution of Carbopol 940: the same as that in [? ?]. A complete rheological characterization was made using a TA instrument AR 2000 controlled torque rheometer. The flow curves (shear-viscosity μ vs shear-rate $\dot{\gamma}$) are very well fitted by the Herschel-Bulkley model for the whole range of shear rates encountered in our experiments. In all our experiments, the shear thinning index $n \approx 0.5$ and the ratio of the radius of the pseudo-plug zone [?] to that of the pipe is less than 0.1.

Results and discussion. As it has been advocated by [?] among others, a reliable indication of the onset and offset of transition is obtained by plotting the turbulence intensity I_t, i.e., the ratio of the root-mean-square of the axial velocity fluctuations $\hat{w}'$ to the bulk velocity $\hat{W}_b$, against the Reynolds number Re_w. Fig. 1(a) displays the evolution of I_t with Re_w at the azimuthal position $\theta = -\pi/4$ and different radial positions. Here, the anti-clockwise orientation is adopted and $\theta = 0$ is the horizontal plane. In the laminar regime and for a given radial position, I_t remains practically constant. At $Re_w = Re_{c1} \approx 1800$, the laminar regime ceases to be a global attractor and a new state, called here, nonlinear asymmetric state, is selected by the fluid. A smooth increase of the turbulence intensity is observed. It reaches a maximum, at $Re_w \approx 3000$, then a plateau or decreases slightly. At $Re_w = Re_{c2} \approx 4000$, a sharp increase in I_t occurs across the pipe section. It reaches a maximum I_{tmax} at $Re_w = Re_{c3} \approx 6500$ and then relaxes to the value corresponding to fully developed turbulence. Two particular stages in the evolution of I_t v.s. Re_w are identified. The first stage corresponds to $Re_{c1} \leq Re_w \leq Re_{c2}$ and the second one to $Re_{c2} \leq Re_w \leq Re_{c3}$. The first stage is not observed for newtonian fluids while the second one is rather "classical" and starts with the appearance of turbulent puffs. In the first stage, the experimental measurements of the friction factor are very close to the theoretical laminar solution as shown in Fig.1(b). Another feature of this first stage is the asymmetry of the mean (time-averaged) flow, a three-dimensional description of which is given in [?]. Axial velocity profiles were measured at three axial positions $\hat{z} = 20D$ (near the entrance section), $\hat{z} = 54\,D$ (middle of the pipe) and $\hat{z} = 122\,D$ along four diameters shifted of 45 degrees. The mean velocity profiles obtained $W(r, \theta, z) = \hat{W}(r, \theta, z)/\hat{W}_b$ are then written as the superposition of an averaged mean axial velocity profile $\overline{W}(r,\, z)$ and a streak $W_s(r,\, \theta,\, z)$. The representation of W_s as a function of θ shows clearly that $W_s(r, \theta, z)$ is well described by the relation: $W_s(r, \theta, z) = A(r, z) \cos(\theta + \phi)$. It is thus possible to draw the contours of iso-W_s in a cross section of the pipe, at each axial position. The result of this procedure is given in Fig.2 (At

FIG. 1: (a) Turbulence intensity $\left(\sqrt{\langle \hat{w}'^2 \rangle_t}/\hat{W}_b\right)$ versus the Reynolds number Re_w at $\theta = -\pi/4$ and different radial positions. The continuous lines through the data points serve as a guide for the eye. (b) Friction factor as a function of Reynolds number. The symbols are the experimental measurements and the continuous line is the theoretical laminar solution.

FIG. 2: Contours of iso-W_s at (a) $\hat{z} = 54\,D, Re_w = 2420$, (b) $\hat{z} = 122\,D, Re_w = 2420$ and (c) $\hat{z} = 122\,D, Re_w = 3650$. The flow is faster in the red zone and slower in the blue zone. The degree of asymmetry evaluated by $max(W_s) \times 100$ is given below each figure.

$\hat{z} = 20\,D$, W_s values are within experiment uncertainty, and are not represented). The red color indicates regions where the fluid-flow in the direction of the pipe is faster than average, while blue denotes regions that are slower. This nonlinear asymmetric state is stable and persists for the whole duration of the experiments (several weeks). These streaks suggest the existence of a coherent structure characterized by two counter-rotating longitudinal vortices. Slow flow is advected from the wall toward the blue zone and fast flow is advected toward the red zone. An example of the velocity-time history signal in the nonlinear asymmetric state is displayed in Fig. 3(a). Frequency power spectra of the axial velocity fluctuations at $r = 0.7$, $\theta = -\pi/4$ (near the center of the high speed streak) for different values of Re_w are shown in Fig. 3(b). These spectra were calculated from stationary time series (of 5×10^5 data points) of the measured axial velocity. The data points are not evenly spaced, so a refined sample and hold technique was used to compute the power spectra [?]. The spectra were calculated with segments of 1024 and 2048 data points and the obtained spectrum is an average over all segments in the full time series. In the beginning of the first stage, part of the energy spectrum scales as $f^{-\alpha}$ with α close to 5/3. With increasing Re_w, the spectra have a broad region of frequencies, where the fluctuation energy decays according to a power law $E \sim f^{-\alpha}$ with α very close to 3. The flattening of the curves at high f is due to instrumental noise. The power-law decay region spans about an order of magnitude in $f : 2 \leq f \leq 10$Hz, which implies excitation of the fluid motion in the whole range of the corresponding temporal scales. Assuming that the Taylor hypothesis of frozen turbulence can be used here, because $rms(\hat{w}')/\hat{W} \leq 10\%$, we can view the spectra in time as spectra in space, with the relation between the frequency and the wave number given by $k = 2\pi f/\hat{W}$. Then the power-law decay regions in curve 5 of figure 3(b) means that the fluid motion is excited in the whole corresponding ranges of $k : 0.695 \leq k \leq 7.2\, m^{-1}$. This multiplicity of spatial and temporal scales is one of the characteristic features of turbulence. It is thus reasonable to suggest that under certain conditions, a truly turbulent flow might be excited by the nonlinear variation of the effective viscosity with the shear rate. Actually, we have an interplay between inertia and shear-thinning. This idea was first put forward by [?]. The large value of the exponent means that the power of fluctuations decays very quickly as the size of eddies decreases. Therefore, the fluctuations of the velocity and the velocity gradient are both determined by the integral scale. The flow can be considered smooth in space and random in time. This situation is analogous to Batchelor regime [?]. An estimation of the characteristic temporal and spatial scales associated to the axial velocity fluctuations can be obtained using the autocorrelation function $C_{ww}(T)$ defined by $C_{ww}(T) = \dfrac{\langle \hat{w}'(t).\hat{w}'(t+T)\rangle_t}{\langle \hat{w}'^2\rangle_t}$, where T is the delay time. In figure 4(a) we report the autocorrelation function $C_{ww}(T)$ for different values of Re_w. One can note that $C_{ww}(T)$ increases with increasing Re_w. At sufficiently large Re_w, a strong correlation, indicated by the level of the plateau, is observed. Using Taylor's frozen flow hypothesis, we may estimate the integral length scale $\hat{L}_c$, i.e., the

FIG. 3: (a) Time serie of axial velocity for $Re_w = 3650$ at $\hat{z} = 122\,D$, $r = 0.7$ and $\theta = -\pi/4$. (b) Density energy spectra of axial velocity fluctuations at $r = 0.7$, $\theta = -\pi/4$ and different Re_w: (1) $Re_w = 2010$; (2) $Re_w = 2161$; (3) $Re_w = 2327$; (4) $Re_w = 2680$ and (5) $Re_w = 3650$. For $3000 \leq Re_w \leq Re_{c2} \approx 4000$ the spectra are very close. We have represented only that at $Re_w = 3650$. The dashed and dotted lines represent the power law behavior respectively f^{-3} and $f^{-5/3}$.

FIG. 4: Statistics of the axial velocity fluctuations near the center of the high-velocity streak ($r = 0.7$, $\theta = -\pi/4$). (a) Autocorrelation function at $\hat{z} = 122D$ and (1) $Re_w = 2010$, (2) $Re_w = 2161$, (3) $Re_w = 2367$, (4) $Re_w = 2420$, (5) $Re_w = 3650$, (6) $Re_w = 2970$. (b) Integral scale as function of the Reynolds number at two axial locations: (1) $\hat{z}/D = 54$ and (2) $\hat{z}/D = 122$

largest scale on which the velocity is correlated. In figure 4(b) we plot the integral length scale, normalized by the diameter of the pipe, as a function of the Reynolds number at two axial locations $\hat{z}/D = 54$ and 122. As for the turbulence intensity in Fig.1(a), the integral length scale increases with increasing Reynolds number, reaches a maximum and saturates. It increases also along the pipe suggesting a stronger correlation.

Deeper insight on this weakly turbulent flow induced by the shear-thinning behavior of the fluid is obtained from the statistics based on the longitudinal velocity difference taken

FIG. 5: (a)Velocity structure functions $S_p(T)$ for $1 \leq p \leq 6$ at $(r = 0.7,\ \theta = -\pi/4)$, $\hat{z} = 122D$ and $Re_w = 3650$. (b) Relative exponent ξ_p/ξ_3 as a function of p at $Re_w = 3650$. The continuous line corresponds to the Kolmogorov scaling.

over the time scale T: $\delta w'(T) = |w'(t+T) - w'(t)|$. The p-th-order structure function or the p-th-order moment of longitudinal velocity difference is defined as $S_p = \left\langle (\delta w'(T))^p \right\rangle_t$. Figure 5(a) displays a set of $S_p\,(T)$. As shown all moments scale as $S_p\,(T) \propto T^{\xi_p}$ in the range corresponding roughly to the power law decay of the velocity spectra. For T greater than the time integral scale, saturation to constant values is observed. Although the scaling range is limited, the exponent ξ_p nonetheless can be extracted from the slopes of the individual curves. In the Kolmogorov theory of 3D fluid turbulence, assuming self-similar statistics, $\langle (\delta w'(T))^p \rangle$ is predicted to scale as T^{ξ_p} with $\xi_p = p/3$. Benzi *et al*[**?**] have introduced a new concept known as extended self-similarity (ESS). According to this concept, the scaling is significantly better if data are presented in the form: $S_p = S_3^{\xi_p^*}$. The deviation from the linear relation, as p gets large [**?**], is attributed to the intermittency of the velocity fluctuations. In the present study, because of the small range where the power-law holds, the ESS process is adopted. The dependence of the normalized scaling exponent ξ_p/ξ_3 on the order of the structure function, p, is presented in Fig. 5(b). The experimental data hardly depart from the Kolmogorov line. We may thus conclude that there is no substantial intermittency.

Attempt of interpretation. The starting point is the standard Reynolds decomposition. The capital letters or an overbar denote time-mean values and small letters with prime designate fluctuating quantities. The Reynolds-averaged momentum equation is:

$$\frac{\partial V_i}{\partial t} + \frac{\partial}{\partial x_k}\left(V_i\, V_k\right) = -\frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_k}\left(\frac{1}{Re}\overline{\mu'\dot{\gamma}'}_{ik} - \overline{v_i'\, v_k'}\right) + \frac{1}{Re}\frac{\partial}{\partial x_k}\left(\overline{\mu}\,\Gamma_{ik}\right),$$

where Γ_{ik} are the components of the strain-rate tensor. By comparison with the newtonian case, there is a new diffusive term $\overline{\mu'\dot{\gamma}'_{ik}}$, which may be called a non-newtonian Reynolds-stress tensor. If one considers the evolution of the turbulent kinetic energy, a new term $I_{\mu'} = -\overline{\mu'\dot{\gamma}'_{rz}}\frac{\partial W}{\partial r}$ is found. It arises from the contraction of the non-newtonian Reynolds-stress tensor with the mean strain-rate tensor. Using first order Taylor approximation for μ', it can be shown easily that $I_{\mu'} = -\overline{\dot{\gamma}'^2_{rz}}\Gamma_{rz}\left(\partial\overline{\mu}/\partial\Gamma_{rz}\right) > 0$, that is to say, it is a source of growth of fluctuating kinetic energy. Therefore, we can speculate that this source term $I_{\mu'}$ contributes to sustaining the weak turbulent flow. This is probably one of the pieces of the puzzle associated to the weakly turbulent flow described here. An other piece could be the merging of small vortices in a cross-section to form a strong vortex dipole. Currently, the link between these points of view is not elucidated.

As a conclusion, we have clearly identified what is called here, the nonlinear asymmetric state. A complete statistical analysis is provided. Experimental evidence of chaotic flow induced by the shear-thinning behaviour is reported. Finally, a non-newtonian Reynolds-stress tensor is defined which could be at the origin of this chaotic flow. *Acknowledgment.* The authors are very grateful to A. Bottaro, J.P. Brancher, G. Homsy and E. Plaut for fruitful discussions.

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