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# Mechanisms of elastic turbulence in gelatinized starch dispersions

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ABSTRACT

The aim of this work is to study the rheological response of gelatinized starch dispersions under constant shear stress. To this end, starch dispersions at four different starch concentrations, were prepared by stirring and heating at 90 °C by 20 min. The experiments showed that the mechanical (i.e., strain) response is composed by a long-term trend that can be described by a two-relaxation mode process, and a high-frequency unstable response. Optical images indicated that the compact packing of the insoluble amylose-rich material, known as ghosts, is responsible for the unstable flow response. In fact, after destroying the starch dispersion microstructure with severe shear conditions (sonication), it was observed that the unstable flow response was no longer present. Fourier and fractal (DFA) analyses showed that the scaling characteristics of the strain instabilities depend on the starch concentration and the applied shear stress value. Also, the characteristic flow curves suggested that yield stress and non-monotonous flow curves are at the center of the mechanisms triggering elastic turbulence in starch dispersions.



## 1. Introduction

It is well known that inertial effects (i.e., momentum) can lead to flow instabilities, such as vortex turbulence and intermittencies. Instabilities appear when a critical Reynolds number is achieved. Since the Reynolds number is related to the ratio of forces caused by inertia over those of the flow resistance of the fluid, so relatively high Reynolds number are related to low viscosity samples. However, experimental results on solutions of flexible long-chain polymers showed that complex flow patterns can arise at relatively low Reynolds numbers where inertial mechanisms play a marginal role in the flow development [1]. The phenomenon, called as elastic turbulence, was characterized by a significant stretching of the polymer molecules and an increased elastic stress of two orders of magnitude. Given the marginal role of inertial effects in the Navier–Stokes equations, the elastic turbulence effect was explained by considering nonlinear constitutive relationships between the elastic stress and the rate of flow deformation [2,3]. Pure elastic flow instabilities under the absence of inertial forces in devices used for viscoelasticity measurements were reported in a detailed review [4].

A diversity of experiments and numerical simulations exploring elastic turbulence has been reported. Experiments include rotational flows between two cylinders [5] and plates [6,7], also in curved channels [8], around obstacles [9], and in long, straight micro-channels [10]. It has been pointed out [10] that most of the nonlinear flow behavior observed in

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elastic turbulence studies can be explained from the elastic stresses due to the mechanical behavior of polymer molecules in the fluid. The underlying elastic stresses evolve on a wide range of time-scales reflecting the time needed for a polymer molecule to relax to its equilibrium state [11]. As a matter of fact, the equilibrium relaxation times of polymeric solutions can become of the order of seconds to minutes [12]. In this regard, elastic turbulence appears in hydrodynamic turbulence below the dissipation scale.

The vast majority of studies on elastic turbulence have been related to highly structured complex fluids, including micellar solutions [13–16], dense lamellar phases [17–20] and dense suspensions [21,22]. Shear banding is commonly found in flow instabilities related to elastic turbulence in wormlike micellar systems [23]. Complex spatio-temporal fluctuations have been attributed to different mechanisms, including wall slip, interfacial instability between bands or bulk instability of one of the bands. It has been shown that type-II intermittence can explain the route to chaos in wormlike micelles with flow-concentration coupling [15]. Also, the observed spatial dynamics of the interfaces of vorticity rolls depended crucially on the gap width of the Couette cell [16]. The reader is referred to a recent critical review of the experimental results and possible instability mechanisms related to wormlike micellar systems [24]. For dense lamellar phases, rheochaos has been explained with the occurrence of structural changes [25].

The idea behind the unstable flow behavior of structured complex fluids is that the fluid components rearrange themselves for adapting to changing shear conditions. Overall, these results suggested that the existence of a complex, highly elastic microstructure is an important ingredient for obtaining rheological instabilities under very-low Reynolds numbers [26]. In turn, it should lead to the manifestation of secondary vortex flows and increased flow resistance [27].

Yet the existence of elastic turbulence in flows of biopolymer solutions has been rarely explored. Biomaterial-based fluids commonly exhibit complex microstructures, including highly elastic fractions. Biopolymers, among other application, are widely used in biomedical, pharmaceutical and food industries as thickeners, thinners, gelation agents, and emulsion stabilizers [28]. Gelatinized starch dispersions are particularly interesting in this regard since starch dispersions are composed of dispersed amylopectin-rich swollen starch particles embedded in a continuous 3D network created by the dissolved amylose [29,30]. Swollen starch particles, known as ghosts because of their appearance in optical microscopy, are highly elastic structures moving freely in a continuous 3D network [31]. It has been shown that starch dispersions can exhibit nonlinear viscoelasticity due to inertial effects [32]. Similar to giant micelle dispersions, these ingredients, in combination with relatively small shear stress conditions, can lead elastic turbulence in rheological measurements. In a recent work, we showed that starch dispersions display chaotic rheological patterns that can be attributed to flow instabilities [33].

The aim of this work was to further exploring the nature and characteristics of elastic turbulence of gelatinized starch dispersions. Whereas in a previous work [32] we reported elastic turbulence for gelatinized starch dispersions at 5% (w/w), in this work we explore the effects of starch concentration. To this end, gels at four different starch concentrations were prepared by cooking starch dispersions at 90 °C for 20 min. Rheological measurements were carried out in a shear stress-controlled rheometer at room temperature (25 °C). The measured shear strain response exhibited complex patterns in relation to different constant shear stress conditions. The analysis of the measured signals by means of Fourier analysis and detrended fluctuation analysis suggest that the flow instabilities have an elasticity-dominance origin.

## 2. Experimental set-up

Four corn starch (Sigma–Aldrich, Saint Louis, MO) concentrations were considered; namely, 3, 4, 5 and 6% (w/w). The reader is referred to our previous work in [32] for details of the starch specifications and the preparation of the gelatinized starch dispersions.

### 2.1. Rheological measurements

The results in this work were based on the determination of the shear strain in the bulk of the starch dispersions in response to a constant shear stress application. The procedure and the equipment used are detailed in our previous work [32].

## 3. Detrended fluctuation analysis

The detrended fluctuation analysis (DFA) [34] has been established as an accurate method to detect such correlations in data affected by trends. The method is based on random walk theory. For completeness in the presentation, a brief description of the method will be given below. In DFA, the time-series $x_k, k = 1, \ldots, N$, is first integrated

$$Y_k = \sum_{j=1}^{k} (x_j - \langle x_k \rangle), \quad k = 1, ..., N \tag{1}$$

where $\langle x_k \rangle = \frac{1}{N}\sum_{j=1}^{N} x_j$ is the time-series mean. After dividing $Y_k$ into $N_s = [N/s]$ not-overlapping segments of equal length $s$, a piecewise polynomial trend $Y_{s,k}$ is estimated within each segment and the detrended series is calculated as $\tilde{Y}_k = Y_k - Y_{s,k}$. The degree of the polynomial fit can vary in order to eliminate linear, quadratic or higher order trends of the integrated time series. Here we use linear polynomials for detrending. The fluctuation function is computed as

$$F(s) = \left( \frac{1}{sN_s} \sum_{j=1}^{sN_s} \tilde{Y}_k^2 \right)^{1/2} \tag{2}$$

Each integrated time series is self-similar if the fluctuation function $F(s)$ scales as a power law with the segment size $s$ (i.e., the number of strides or complex values in a segment of observation or time scale). Typically, $F(s)$ increases with the segment size $s$. A linear relationship on a double-log graph indicates that

$$F(s) \approx s^{\alpha} \tag{3}$$

where the scaling exponent $\alpha$ (also called the self-similarity or self-affine parameter) is determined by calculating the