

*Rapid Note***Faraday instability with a polymer solution**F. Raynal¹, S. Kumar², and S. Fauve^{2,a}¹ Laboratoire de Mécanique des Fluides et d'Acoustique^b, École Centrale de Lyon et Université Lyon I, 36 avenue Guy de Collongue, B.P. 163, 69131 Écully Cedex France² Laboratoire de Physique Statistique^c, École Normale Supérieure, 24 rue Lhomond, 75005 Paris, France

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Abstract. We present an experimental study of the Faraday instability in which we compare the behavior of a Newtonian fluid (water-glycerine mixture) with that of a semi-dilute non-Newtonian solution of high molecular weight polymer. We show that although the dispersion relation of surface waves, derived for a layer of inviscid fluid, remains valid in that particular non-Newtonian case, the behavior of the instability threshold with frequency strongly differs from the Newtonian case. We explain this effect as a result of a frequency-dependent viscosity. The linear stability analysis of the non-Newtonian case shows a perfect agreement with the experimental results both for the dispersion relation and for the reduction of the instability threshold. We discuss the use of the characteristics of the Faraday experiment as a measurement tool to determine frequency dependent properties of non-Newtonian fluids.

PACS. 47.20.-k Hydrodynamic stability – 47.50.+d Non-Newtonian fluid flows

Parametric amplification of surface waves by vertically vibrating a horizontal layer of fluid has been widely studied since Faraday [1]. Most experiments have been performed with Newtonian fluids and motivated by the study of pattern forming instabilities and chaos. However, it has been shown that the instability characteristics also provide a simple way to determine the fluid properties (surface tension, viscosity) even when other measurement techniques are difficult to handle (in the vicinity of a liquid-vapor critical point) [2,3]. We present in this note a similar study, but with a liquid layer of polymer solution. We show that the measurements of the instability wavelength and of the critical acceleration of the container for instability onset determine rheological properties of this non-Newtonian fluid. We observe that the dispersion relation of surface waves on an ideal fluid layer allows a good fit of the experimental data for the wavelength measured in the polymer solution. On the other hand, measurements of critical acceleration indicate that the viscosity of the polymer solution decreases with increasing frequency. These results are confirmed by a linear stability analysis. Although frequency-dependent viscosities are well documented in polymer rheology [4], we discuss the particular characteristics of our flow configuration and conclude on

the use of the Faraday instability as an experimental tool in polymer rheology.

The experimental set-up has been described in details elsewhere [5]. The vibrations are generated using a shaker table (Brüel & Kjær type 4808), powered by an audio amplifier (Brüel & Kjær 2712), itself fed by the out sine wave of a multifunction Synthesizer (HP 8904A). The threshold acceleration is measured using a piezoelectric accelerometer (Brüel & Kjær 4375) connected to a charge amplifier (Brüel & Kjær 2635).

The container is an aluminum-made parallelepiped, with dimensions $L_x = 140$ mm and $L_y = 13$ mm in length and width, and $h = 11$ mm in depth. We used the brimful boundary condition[6]. The fluid surface is completely flat before the instability onset.

A good temperature control is necessary in these experiments since viscosity changes rapidly with temperature. In order to achieve this condition, the cell is specially designed with a water circulation, thermally regulated with a waterbath set to 14 °C within 0.1 °C (this temperature is chosen in order to avoid water evaporation). The cell is closed with a plexiglass cover which avoids heat losses, prevents the fluid from evaporating and overall, avoids the gathering of contaminants at the interface, which would cause a change of surface tension. The fluid temperature, measured *in situ* slightly above onset (so that mixing reduces temperature gradients across the cell), is thus equal to 14.5 °C.

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Special care is taken in order to get a perfectly homogeneous pattern at instability onset. To wit, we use a carefully balanced cell, the horizontal extent of which is smaller than the shaker table in order to avoid sloshing motions.

The experiments are performed for frequencies ranging from 40 Hz to 260 Hz at most. Indeed, below 40 Hz, the number of wavelengths in the large direction, equal to 12.5 for this frequency, rapidly decreases so that the wavelength becomes too large and quantization effects by lateral boundaries are apparent. The upper frequency limitation is related to the power amplifier which cannot drive the cell at accelerations larger than roughly 15 g. The instability onset can thus be observed up to 200 Hz with the water-glycerine solution, and 260 Hz with the polymer solution (see Fig. 3).

The Newtonian solution is a water-glycerine mixture made with distilled water. Its exact kinematic viscosity was measured with a Ubbelohde apparatus in a cooling bath at temperature 14 °C: we found $\nu = 5.9 \times 10^{-5} \text{ m}^2/\text{s}$, *i.e.* a dynamical viscosity $\mu = \rho\nu = 6.6 \text{ cp}$, where ρ is the volumic mass of the mixture, measured to be $\rho = 1.12 \times 10^3 \text{ kg/m}^3$ at this temperature. The non-Newtonian solution is a semi-dilute polymer solution in water, with concentration $C = 5 \text{ g/l}$. It is made homogeneous by heating from below up to 50 °C and slowly agitating for 24 hours. The polymer used is a long-chained polydisperse polyacrylamide (Aldrich No. 18, 127-7), with average molecular weight $M_w = 5 \times 10^6 \text{ g/mol}$. This corresponds to an average number of monomers $N \sim 7 \times 10^4$.

We first studied the dispersion relation, which for a layer of inviscid fluid with depth h and an infinite horizontal extension is given by:

$$\omega_0 = \sqrt{\left(gk + \frac{\sigma}{\rho}k^3\right) \tanh kh} \quad (1)$$

where $g = 9.81 \text{ ms}^{-2}$ is the acceleration of gravity, k is the wavenumber, ω_0 is the wave pulsation, and σ is the surface tension. It has been shown that it predicts rather accurately the instability wavelength for Newtonian fluids, even one hundred times more viscous than water [5, 7]. In our parameter range, the wave generated by the instability is subharmonic, thus $\omega_0 = \omega/2$, where ω is the pulsation of excitation. In a parallelepipedic cell, the wavenumber k is given by the following formula:

$$k = 2\pi \sqrt{\left(\frac{n_x}{L_x}\right)^2 + \left(\frac{n_y}{L_y}\right)^2}, \quad (2)$$

where n_x and n_y are the number of wavelengths in the x and y direction. Note that because of the brimful boundary condition, n_x and n_y are necessarily half integers or integers (the linear modes are standing waves with a zero-amplitude on the boundaries in the two directions). In the case of our elongated cell, there is only half a wavelength in the y -direction (in the range of frequency studied), thus $n_y = 1/2$. The wavenumber k is therefore very easily calculated by counting the number n_x of wavelengths in the

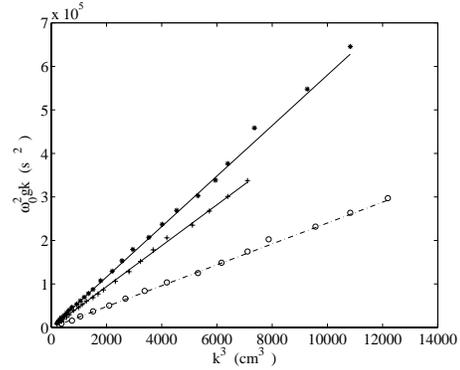


Fig. 1. The quantity $\omega_0^2 - gk$ where ω_0 is the pattern pulsation and k its wavenumber, is plotted as a function of k^3 for differently contaminated polymer solutions. Experiments: (*), polymer with distilled water; (+), polymer with tap water; (o), polymer solution with teepol. Linear theoretical fits: (—), $\sigma = 58 \text{ dyn/cm}$; (---), $\sigma = 47 \text{ dyn/cm}$; (- · -), $\sigma = 24 \text{ dyn/cm}$.

x -direction, and using equation (2). Note that the depth h is sufficiently large so that in the range of studied frequencies, $\tanh kh \simeq 1$.

This relation is first checked with silicone oil (Rhodorsil 47 V 20) at room temperature of known and robust surface tension (20.5 dyn/cm at 25 °C). Using the detachment method [8], we measure $\sigma = 21.7 \text{ dyn/cm}$. Using the dispersion relation, the best fit obtained with equation (1) from the experimental data is $\sigma = 23.4 \text{ dyn/cm}$. This value is roughly 10% larger than the one measured using the detachment method. In the case of an inviscid fluid, it has been shown that the edge constraints modify the dispersion relation and thus can explain this difference [6, 9]. This difference disappears in the case of large aspect ratio containers [5].

Then we consider three solutions of polymers as described before, one in distilled water, the second one in tap water, and the third saturated with teepol, a surfactant.

In our parameter range, $\tanh kh \simeq 1$. Thus, we plot $\omega_0^2 - gk$ versus k^3 in Figure 1: the dots correspond to the experimental points (the error is about of the size of the dots), and the full lines are the linear best fits, from which one can obtain the surface tension σ for each solution. The linear relation between those two quantities shows that the gravity and capillary forces are dominant with respect to the elastic or viscous ones in the explored frequency range [10]. Note that the attempts to measure the surface tension for the polymer solutions with the detachment method gave values far above those obtained with the dispersion relation (due to migration of polymers from the solution to the platinum plate). Note also that we did not try here to look at the dependence of surface tension versus polymer concentration, but the Faraday instability provides a simple way of performing such a study. Nonlinear effects might be also considered by looking at regimes far from the instability threshold.

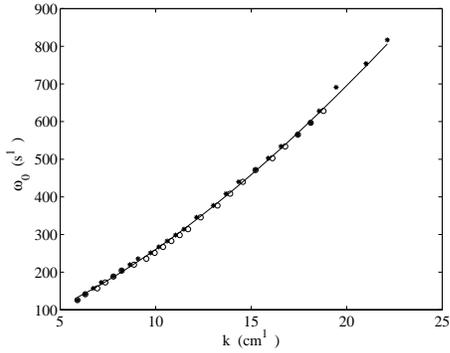


Fig. 2. Pattern pulsation ω_0 as a function of the wavenumber k : (\circ), water-glycerine solution; ($*$), polymer solution; (—), theoretical fit with $\sigma = 58$ dyn/cm.

We next consider the critical acceleration for instability onset. Our aim is to obtain a Newtonian fluid and a polymer solution with the following properties:

- (1) the same dispersion relation in the explored frequency range, (*i.e.* the same surface tension),
- (2) the same acceleration threshold for instability onset at low frequency (40 Hz).

Proceeding this way, a good comparison can be obtained between the Newtonian and non-Newtonian behaviors of the instability threshold with respect to the excitation frequency. The threshold obtained with the water-glycerol solution can be easily changed by varying the concentration of glycerol. The surface tension of both solutions can be varied by adding contaminant. We tried a great number of different solutions and were able to achieve the above conditions. For each frequency, two measurements of the acceleration are performed, one just below onset when the interface is flat, the other just above onset, when patterns are present. The acceleration a shown in Figure 3 is the mean of the two values, while their difference gives the domain of uncertainty: a is given within ± 0.015 g. Note that no hysteretic effects were detected with the polymer solution when raising the acceleration above the threshold and then going back to smaller accelerations. Figures 2 and 3 show the dispersion relation and the amplitudes of the respective thresholds for the Newtonian and non-Newtonian fluids: as can be seen, although the two solutions have exactly the same surface tension $\sigma = 58$ dyn/cm, and give nearly the same instability threshold at 40 Hz, the instability thresholds at higher frequencies strongly differ: the threshold grows less rapidly in the non-Newtonian case. The instability threshold corresponds to the balance between excitation by periodic forcing and viscous dissipation. It is an increasing function of viscosity. All the other parameters of the problem being identical in the experiments with the Newtonian and non-Newtonian fluids, we conclude from Figure 3 that the viscosity of the polymer solution is a decaying function of frequency.

In order to understand the results of these experiments, a linear stability analysis has been performed using

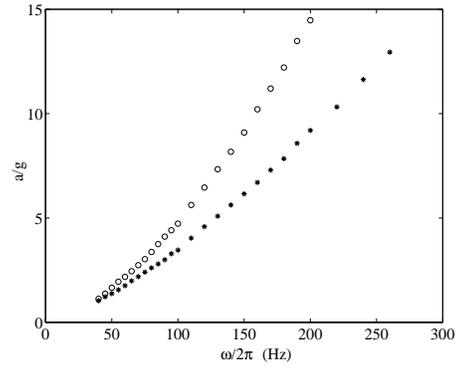


Fig. 3. Threshold amplitude Ac/g as a function of frequency of excitation $\omega/2\pi$: (\circ), water-glycerine solution; ($*$), polymer solution.

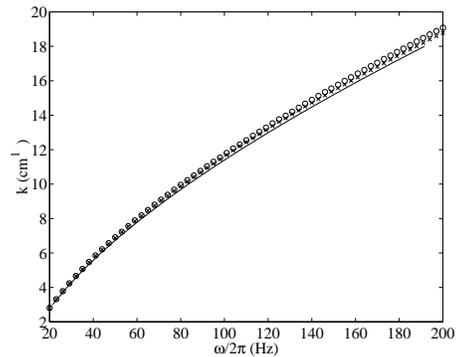


Fig. 4. Calculated dispersion relations for the fluids used in these experiments. The circles correspond to the Newtonian fluid and the crosses to the non-Newtonian fluid. The solid line is the calculated dispersion relation for an inviscid fluid of the same surface tension and density.

a single-mode Maxwell model to describe the fluid rheology [4]. Detailed results of this analysis will be reported elsewhere [11], but here we present some preliminary findings which are relevant to the present experiments. In Figure 4, we show the dispersion relations calculated using parameters which correspond to the Newtonian and non-Newtonian fluids of the experiments, as well as the dispersion curve for an inviscid fluid of the same surface tension and density. For the non-Newtonian fluid we have assumed a relaxation time of 5×10^{-3} s, which is a reasonable value for a semi-dilute polymer solution [10]. All three dispersion relations lie along approximately the same curve, consistent with experimental findings. Interestingly, the critical wavenumbers for the Newtonian and non-Newtonian cases are slightly larger than the inviscid values, indicating that the presence of relatively weak viscous forces does not necessarily make larger lengthscales more favorable. (As the zero-shear viscosity increases, the Newtonian and non-Newtonian wavenumbers do become smaller than the inviscid values, as is expected.) In Figure 5, we show the threshold curves for the same Newtonian and non-Newtonian solutions. The results are in perfect qualitative agreement with the experiments: the thresholds are

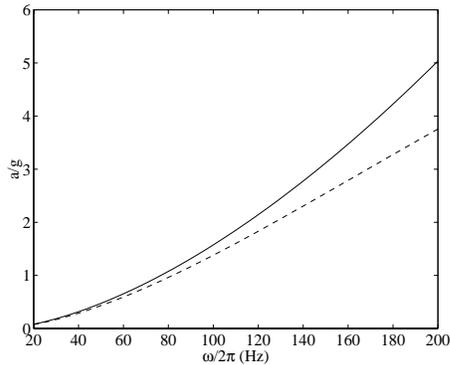


Fig. 5. Calculated thresholds for the fluids used in these experiments. The solid line corresponds to the Newtonian fluid and the dashed line to the non-Newtonian fluid.

the same at low frequency, but the non-Newtonian threshold grows less rapidly as the forcing frequency increases. This behavior can be explained by noting that a Maxwell fluid has a frequency-dependent complex-valued viscosity in an oscillatory flow [4]. The real part of this viscosity is proportional to the energy dissipation and decreases as the frequency increases, resulting in lower thresholds. Note that the calculated thresholds for both the Newtonian and non-Newtonian cases are lower than the experimental values by about a factor of 3. A closer look shows that this ratio decreases by roughly 30% as the frequency is increased from 40 to 200 Hz. This shows that, as expected, the finite size effects become smaller when the wavelength decreases, and strengthens the idea that the higher experimental values are most probably due to the strong damping at the lateral walls, which is not taken into account by the calculations [12–14]. The elongated geometry was motivated by the experimental simplicity for determining the wavenumber. We anticipate that experiments performed in a wider container will show much better quantitative agreement.

Our experiment has given the following information:

- (1) this instability wavelength characterizes the nature of the surface mode (capillary or elastic) of the polymer solution and gives the value of the surface tension. The Faraday instability could be used in the same way to investigate the dependence of surface tension on the polymer concentration in the capillary regime or to study the elastic regime;
- (2) the behavior of the instability threshold with frequency strongly depends on the concentration of polymer. At high enough concentration, this behavior indicates that the viscosity of the polymer solution decreases with increasing frequency.

Note that at the Faraday instability onset, the shear is null (slightly above onset, it can be made as small as possible). The basic flow is thus just an oscillatory flow. This experiment with a non-Newtonian fluid thus strongly differs from those related to the Taylor-Couette instability

(instability between two rotating cylinders) [15,16], for which the basic flow is stationary but with a non-zero shear.

Different non-Newtonian effects are probed with these two instabilities.

Finally, we would like to comment on the possible use of the Faraday instability as a measurement tool in polymer rheology. As already mentioned, the linear stability analysis performed with one of the available models used in rheology will allow the determination of the frequency-dependent viscosity from the measurement of the instability thresholds as a function of frequency. This measurement method has the following advantages.

- (1) Its range of frequency completely differs from that of classical rheometers (generally less than 15 Hz). The 250 Hz upper limit of the present study could be easily increased by using a more powerful vibration exciter. There is no filtering effect due to the inertia of a mechanical measuring device.
- (2) There are no boundaries in relative motion. Therefore no measurement errors due to slipping of the polymer at a moving boundary could be made.
- (3) Only linear terms of the fluid equation are probed at instability onset.

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