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# Dynamic contact angle measurements of viscoelastic fluids

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# ABSTRACT

In this study, the dynamic contact angles of a series of viscoelastic fluids were measured through a modified Wilhelmy plate technique. The advancing and receding contact angles were measured by immersing and withdrawing a PTFE (Teflon) Wilhelmy plate from a reservoir containing a series of different Newtonian and viscoelastic test fluids. The viscoelastic fluids that were tested consisted of either solutions of polyethylene oxide or polyacrylamide in water where the concentration of the polymer was varied to produce solutions with different amounts of fluid elasticity with and without shear thinning. The advancing contact angles of all the viscoelastic fluids tested were found to increase with increasing plate velocity. Conversely, the receding contact angles in each case were found to decrease with increasing contact line velocity. A number of previous measurements have been performed for shear thinning fluids. The measurements presented here are the first to probe the response of the dynamic contact angle at large Weissenberg numbers where the elasticity of the liquid becomes important. For fluids with increased fluid elasticity, the onset of the variation of receding contact angles was found to be delayed to higher contact line velocities and capillary numbers. It was also found that, unlike the case of Newtonian fluids, the cube of advancing and the cube of the receding contact angles were both found to be proportional to the square of the capillary number for highly elastic fluids. Finally, a simple model was proposed to account for the role of elasticity and shear thinning of the viscosity on the dynamic contact angle.

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## 1. Introduction

The dynamic wetting of a liquid along a solid surface is a phenomenon that occurs in great number of natural and industriallyrelevant processes. Over the last few decades, there has been a great deal of research performed, both experimental and theoretical, with the goal of better understanding the dynamic wetting processes over a wide range of flow conditions, and fluid and solid properties [1–4]. Still, there are a number of open questions that remain. In this paper, we will investigate the effect that fluid rheology and specifically viscoelasticity can have on dynamic wetting. We will demonstrate that the addition of fluid elasticity can modify the contact line dynamics and have a great influence on the evolution of contact angle with contact line velocity.

In order to quantify the effect of viscoelasticity on the wetting dynamics of a liquid on a non-deformable solid surface, the shape of the fluid interface can be measured along with the resulting contact angle made between the fluid and the surface. At rest, the contact angle can exist anywhere between the advancing and receding contact angles [1]. For a Newtonian fluid under flow, molecular-level adsorp-

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tion/desorption processes and macroscopic viscous dissipation can result in an increase in the measured advancing contact angle and a decrease in the measured receding contact angle beyond its static value [1,2]. The value of the contact angle for a moving three phase contact line is thus not fixed, but is dynamic and, depends in a known way on the velocity of the contact line.

Through experimental measurements and theoretical development, the dependence of dynamic contact angles on the speed of moving contact line has been revealed for both wetting and nonwetting Newtonian fluids [2] as well as a limited subset of shear thinning and weakly elastic fluids [5,6]. From these results, two different classes of dynamic wetting models have been developed; molecularkinetic models [7] and hydrodynamic models [8-10]. For the hydrodynamic models, viscous dissipation in the vicinity of the contact line has been shown to lead to a deformation of the fluid interface and a change in the contact angle that scales like  $\theta_D^3 - \theta_S^3 \propto Ca$ . This result is known as the Cox-Voinov-Tanner scaling law. Here  $\theta_D$  is the dynamic contact angle,  $\theta_S$  is the static contact angle and  $Ca = \eta U/\sigma$  is the capillary number where *U* is the velocity of the contact line,  $\eta$  is the viscosity and  $\sigma$  is the surface tension. Although it is widely accepted, the Cox-Voinov-Tanner scaling law is not universal. It has been observed to fail at extremely low capillary number regime,  $Ca < 10^{-4}$ , and at very large capillary numbers Ca > O(0.1), where air entrainment, complete coating and the

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effects of inertia have been observed [11]. In the case of molecularkinetic models, the energy dissipation by a contact line friction resulting from adsorption and desorption of molecules along the moving contact line is responsible for contact angle changes. In the model of Blake and Haynes [7], the dynamic contact angle varies as  $\theta_D = \cos^{-1}[\cos\theta_S - (2k_BT/\sigma\delta^2)\sinh^{-1}(U/2K_\omega\delta)]$  where  $k_B$  is the Boltzmann constant, *T* is the temperature,  $\delta$  is adsorption distance of molecules, and  $K_{\omega}$  is frequency of molecular displacements.

The common forced wetting techniques used to measure the dynamic contact angles are a capillary tube [3,12,13] and a plunge tank [14–17]. For example, using a glass capillary tube, Hoffman [3] measured the dynamic contact angle by displacing Newtonian liquids in the tube filled with air. Petrov et al. [18] and Blake [16,19] measured dynamic contact angles by plunging a solid surface into a tank of liquid. Using a plunge tank, a Wilhelmy plate or a cylindrical strand of material can be immersed or withdrawn from a reservoir allowing the observer to investigate both advancing and receding contact angles. Additionally, in this technique, the effect of gravity and inertial can typically be neglected and the observer can gain access to the variation of the contact line through high resolution, high speed imaging. For these reasons, we chose the Wilhelmy plate technique to make the dynamic contact angle measurements presented here.

The dynamic wetting of non-Newtonian fluids is of great interest because of its application to a great number of industrial applications involving coating flow. That said, there are only a limited number of papers in the literature investigating dynamic wetting of non-Newtonian fluids, none of which probe the range of Weissenberg numbers, where elasticity becomes dominant,  $Wi = \dot{\gamma}\lambda > 1$ . Here  $\dot{\gamma}$ is shear rate and  $\lambda$  is relaxation time of the fluid. Seevaratnam et al. [20] studied aqueous solutions of xanthan gum with molecular weight of  $2 \times 10^6$  g/mol. At the contact line speeds they studied, the response of their fluids was dominated by shear thinning with negligible impact of elasticity. Shear thinning was shown to reduce the viscous bending of the air-water interface near the contact line. As a result, a weak dependence of the dynamic contact angle on capillary number was found which did not following classic hydrodynamic model,  $\theta_D^3 \propto Ca$ . Prior of the work of Seevaratnam et al. [20], Carre and Eustache [21] studied spreading dynamics of shear thinning fluids in wetting and de-wetting modes and generalized the classic hydrodynamic model to shear thinning non-Newtonian fluids. They revealed that the dynamic contact angles of power law shear thinning fluids does not follow the classic hydrodynamic model, rather it depends on the spreading rate to the shear thinning power law exponent *n* such that,  $\cos \theta_s - \cos \theta_D = \frac{K}{\gamma} (\frac{U}{\theta_D})^n$ . Like Carre and Eustache case, the classic hydrodynamic theory was normally applied to describe the dynamic wetting of non-Newtonian fluid. More recently, Liang et al. [22] developed a model to explain the wetting behaviors of non-Newtonian fluids based on Blake's molecular-kinetic theory. Wei et al. [5] tested Boger fluids which are dilute polymer solutions dominated by elasticity and with negligible shear thinning. They found that the curvature of the advancing air-liquid interface was enhanced, but that the capillary number dependence of the dynamic contact angles was not altered compared to the Newtonian fluids. This is likely because the Weissenberg numbers reached in their experiments were all less than one and as a result large elastic effects are not expected.

In this study, we will present dynamic contact angle measurements of a series of viscoelastic fluids. A hydrophobic surface was used so that both dynamic advancing and receding contact angle measurements can be made. The Wilhelmy plate technique allows us to reach high speed of a testing substrate, making it possible to probe the variation of dynamic contact angles over a wide range of capillary numbers. The test fluids used consists of an extremely high molecular weight aqueous polyacrylamide solution with a relaxation time large enough to make high Weissenberg number experiments



Fig. 1. Steady shear viscosity measurements of the test fluids as a function of shear rate. The data include: 0.01 wt% PAM solution (square): 0.05 wt% PAM solution (triangle); viscoelastic PEO solution (star); and Newtonian PEO solution (circle).

possible. We will demonstrate that the dynamic contact angle data at high Weissenberg number do not follow either the expected hydrodynamic or molecular-kinetic scaling even if shear thinning of the fluid viscosity is accounted for. Instead, we will show a much stronger dependence on contact line velocity resulting from the presence of significant elastic stresses in the fluid.

### 2. Description of the experiment

#### 2.1. Materials

A series of test fluids were used for this experiment. For Newtonian fluid pure water was used. In addition, polyethylene oxide (PEO, Sigma Aldrich) with 2  $\times$  10  $^4$  g/mol was used to increase the viscosity of the water without making it viscoelastic. Hereafter, this solution will be designated as Newtonian-PEO solution. For the viscoelastic fluids used in these experiments, two different water-soluble polymers were used. The first was a commercially-available polyacrylamide (PAM) with an extremely high molecular weight (Flopaam 3630, SNF Floerger<sup>®</sup>) often used in enhanced oil recovery. Solutions with two different concentrations of PAM (0.01 wt% and 0.05 wt%) were used in these experiments. As will be shown by detailed rheological measurements, each of the PAM solutions has a large zero shear rate viscosity that shear thins with increasing shear rate, significant fluid elasticity and a large easily-measured relaxation time. A second viscoelastic fluid was tested consisting of an aqueous solution 20 wt% of 2  $\times$  10<sup>4</sup> g/mol PEO and 0.1 wt% of a high molecular weight (8  $\times$  10<sup>6</sup> g/mol) PEO. This Boger fluid was designed to have significant elasticity without shear thinning. Hereafter, this solution will be designated as the viscoelastic PEO solution. All polymer solutions were prepared in deionized water by mixing gently for at least 24 h at a room temperature to obtain a homogeneous solution. The surface tension of the PEO solution was measured to be 0.06 N/m using a pendant drop experiment and the surface tension of both PAM solutions was measured to be 0.07 N/m.

The steady shear viscosity of each solution was measured using a stress-controlled rotational rheometer (TA, DHR3) with a cone-andplate geometry. The results are plotted as a function of the applied shear rate in Fig. 1. As seen in Fig. 1, the viscosity of the viscoelastic PEO solution was found to be constant at  $\eta = 0.088$  Pa s, while the Download English Version:

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