



Dynamic contact angle of dilute polymer solution drops impacting on a hydrophobic surface



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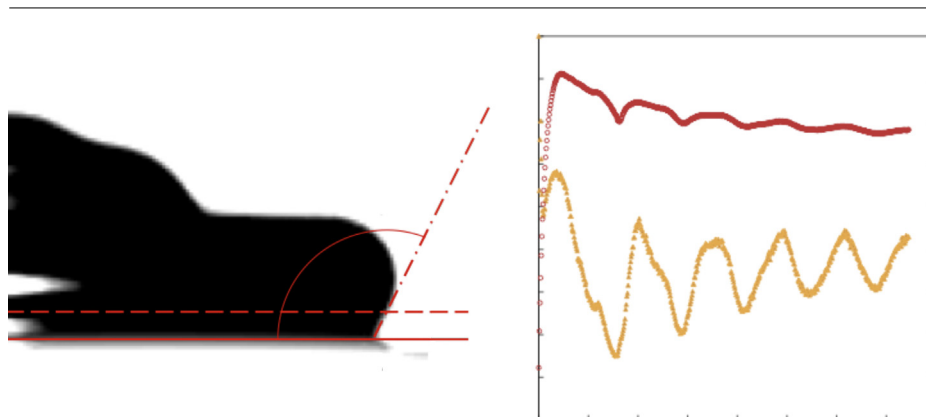
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HIGHLIGHTS

- Systematic measurements of the apparent dynamic contact angle of dilute polymer solution drops are presented.
- Polymer concentrations cover almost entirely the dilute regime.
- Drop impacts onto commercial PTFE surfaces are investigated at different Weber numbers.

GRAPHICAL ABSTRACT



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ABSTRACT

The dynamic contact angle of dilute polymer solution drops impacting on a hydrophobic PTFE surface is studied experimentally by high-speed imaging, for polymer concentrations ranging between 0 ppm and 400 ppm, and impact Weber numbers between 15 and 115. Images with spatial resolution of 16.4 $\mu\text{m}/\text{pixel}$ were captured at 8000 fps to resolve the short time-scale dynamics. Results show that the receding contact angle of dilute polymer solution drops is significantly smaller than in case of drops of pure water, suggesting that the receding contact line encounter a higher resistance to its displacement. The minimum contact angle decreases with respect to both increasing polymer concentration and increasing Weber number. At lower Weber numbers, contact angle oscillations were observed, which can be related to the liquid flow to and from the rim during drop retraction.

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1. Introduction

The equilibrium shape of a drop deposited on a surface is generally described using the well-known Young-Laplace equation with

respect to the interfacial tensions between the solid and the liquid, γ_{SL} , the solid and the vapour, γ_{SV} , and the liquid and the vapour, γ :

$$\gamma_{\text{SL}} + \gamma \cos \theta_{\text{eq}} = \gamma_{\text{SV}} \quad (1)$$

where θ_{eq} is the thermodynamic equilibrium contact angle between the drop and the surface [1,2].

If the drop moves parallel to the surface, however, one can observe an advancing (θ_{A}) and a receding contact angle (θ_{R}), respectively at the leading edge and at the trailing edge. This implies an

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Table 1
Fluid properties.

	0 ppm	50 ppm	100 ppm	200 ppm	300 ppm	400 ppm
$\rho_{20\text{ }^\circ\text{C}}$, kg/m ³	1000	1000	1000	1000	1000	1000
$\mu_{20\text{ }^\circ\text{C}}$, mPa s	1	1.058	1.115	1.23	1.345	1.46
$\sigma_{20\text{ }^\circ\text{C}}$, mN/m	72	70	70	70	70	70
$\tau_{20\text{ }^\circ\text{C}}$, ms	-	15.31	21.65	30.62	37.51	43.31

adhesion force, which can be described either by using Furmidge's equation [3,4]

$$F = \gamma D(\cos\theta_R - \cos\theta_A) \quad (2)$$

where D is the drop base diameter perpendicular to the contact angle plane, or by introducing the concept of line tension [5]:

$$F = \gamma \frac{D}{2}(\cos\theta - \cos\theta_{\text{eq}}) \quad (3)$$

where θ is the apparent (observed) contact angle. A comparative analysis of these two approaches can be found in [6]. Different approaches must be used in case of chemically heterogeneous [7,8] or rough surfaces [9,10], and in case of non-circular drops [11].

An important case where advancing and receding contact angles are observed is drop impact. When a liquid droplet impacts onto a solid surface, it spreads radially taking the shape of a disk (often referred to as "lamella"), eventually surrounded by a toroidal rim. During this stage, the impact kinetic energy is partly converted into surface energy, and partly lost because of viscous dissipation. After reaching maximum spreading, i.e. when nothing is left of the initial kinetic energy, the lamella initiates retracting under the action of capillary forces, in order to minimise surface energy. During the expansion and retraction stages one can observe respectively an advancing and a receding contact angle, which in turn allow the definition of a contact angle hysteresis around the point of maximum spreading.

Whilst the open literature reports several experimental and theoretical studies about the dynamic contact angle of Newtonian drops [12–18], many applications involve the use of complex fluids, such as polymer or surfactant solutions, which exhibit a wetting dynamics remarkably different from the case of simple liquids. Even in the case of very dilute solutions, comparison with the Newtonian solvent (e.g., water) reveals significant differences in the behaviour of the moving contact line during the spreading and/or receding phase, in the amplitude of the dynamic contact angle, as well as in the intrinsic time of the phenomenon [19–21]. A well-known example is the so-called anti-rebound effect of polymer additives: When a droplet of water falls on to a hydrophobic surface, such as the waxy leaf of a plant, the drop is often observed to bounce off, however the addition of very small quantities (~100 ppm) of a flexible polymer can completely prevent rebound, by reducing the recoil velocity of the drop after the inertial spreading of two orders of magnitude [22,23]. This apparently surprising phenomenon (since the shear viscosity and surface tension of such drops are almost identical to those of pure water), is intimately related to the contact angle dynamics, as initially suggested by comparison with drop impact experiments where wetting was artificially reduced or removed [24,25].

Despite their scientific and industrial importance, to date there is a lack of dynamic contact angle data for non-Newtonian drops in the open literature [19]. The present work contributes to address this gap through systematic dynamic contact angle measurements of polymer solution drops impacting on a hydrophobic surface of commercial interest (PTFE), for different impact velocities and polymer concentrations that cover the entire dilute regime.

2. Materials and methods

2.1. Fluids characterisation

The fluids considered in the present work are dilute solutions of polyethylene oxide (PEO) in de-ionized water. The PEO, supplied by Aldrich Chemicals under the form of granular powder, has average molecular weight $M_w = 4000$ kDa, and typical density of 1210 kg/m³. In dilute solutions, the average distance among polymer molecules is larger than their size, so that their interactions are negligible: polymers exhibit a random coil conformation and can be described as spherical particles suspended in the solvent. For a critical value of concentration (the overlap concentration) polymer chains become randomly entangled, which corresponds to a marked increase in the polymer solution viscosity. The overlap concentration can be calculated as:

$$c^\circ = \frac{1}{[\eta]_0} \quad (4)$$

where $[\eta]_0$ is the characteristic viscosity, which for a PEO solution is related to the molecular weight as [26]:

$$[\eta]_0 = 0.0125M_w^{0.78} \quad (5)$$

Thus, for the solutions considered in the present work the overlap concentration is 567 ppm. To investigate the fluid in the dilute regime, one master solution was prepared with a mass concentration of 400 ppm, which was then diluted to obtain solutions at 300, 200, 100 and 50 ppm. Polymer solutions were prepared from a same batch of polymer, protected from direct illumination and from heat sources, and used within 2–3 weeks.

Viscosities were measured by means of a rotational rheometer (Haake MARS II) equipped with a 60 mm plate/plate geometry and Peltier temperature controller, while equilibrium surface tensions were measured using a maximum bubble pressure instrument (Krüss PocketDyne). Finally, the relaxation time was calculated as a function of the characteristic viscosity, concentration, and temperature using the empirical formula [27]:

$$\tau = (1.82 \times 10^{-3}[\eta]_0 - 2.9 \times 10^{-11}[\eta]_0^3 - 0.51)\sqrt{c}\exp(-0.0004T^2) \quad (6)$$

Table 1 summarizes the properties of fluids and drops considered in these experiments. Note that while viscosity is approximately a linear function of the polymer concentration in the dilute regime, the surface tension does not change because PEO saturates the free surface already at low concentrations.

2.2. Experimental setup and procedure

Drops were generated using a syringe with blunt hypodermic needle (gauge 21, 0.495 mm i.d.) driven by a micrometric screw, and detached under their own weight. The characteristic drop diameter calculated from drop weight measurements, $D_0 = (6m/\pi\rho)^{1/3}$, where m is the drop mass, and ρ the water density, was 3.09 ± 0.1 mm for water drops and 2.93 ± 0.1 mm for polymer solution drops; thus, the drop equilibrium radius, $D_0/2$, is smaller

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