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Temperature-dependent soft wetting on amorphous, uncrosslinked polymer surfaces[†]

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The wetting of polymer melts at high temperatures is studied by placing a glycerol drop on a poly (*n*-butyl methacrylate) film and measuring the wetting ridge. The height of the wetting ridge grows continuously over time. These wetting ridge growth rates can be explained by polymer chain dynamics occurring at the molecular level, determined using oscillatory shear rheology of the polymer melt. The shape of wetting ridge profile can be modeled using an equation previously used for elastomers, with a simple modification that incorporates the time-dependent storage modulus of the uncrosslinked melts.

Introduction

The wetting of soft surfaces is important for a range of applications, from polymeric coatings to soft and bioinspired adhesives.¹⁻⁴ Over half a century ago, Lester suggested that the vertical component of surface tension needs to be considered for wetting on soft deformable solids.⁵ This vertical force generates a wetting ridge at the three-phase contact line (TPCL), which can play an important role in drop sliding,⁶⁻¹⁰ coalescence,¹¹ impact,¹²⁻¹⁶ spreading,¹⁷⁻²⁰ and evaporation.^{21,22} Since then, significant efforts have been put forth to describe how a drop wets a soft surface.²³⁻²⁵ However, a full understanding of soft wetting remains incomplete, especially for time- and temperature-dependent situations. The geometry of the wetting ridge is typically governed by a balance of surface/interfacial tensions and substrate elasticity.²⁶⁻²⁸ The vertical peak height of the wetting ridge is known to scale as $h \sim \gamma \sin \theta$, where γ is the surface tension of the drop, θ is the contact angle, and G is the shear modulus of the underlying substrate. Detailed expressions that predict the shape of the wetting ridge profile follow the same scaling.^{19,29} However, this has been mostly verified in static wetting of crosslinked elastomers, and the shape of a growing wetting ridge on an amorphous polymer melt has not been well-described.

When considering the dynamic growth of wetting ridges, viscoelastic properties of the substrate play a role. For example, the growth of an elastomeric wetting ridge around a static drop is related to the viscoelasticity of the solid surface.^{30,31} For the case of a water drop sliding on a soft elastomer, a combination of critical contact angles and rheological properties govern the drop dynamics.^{3,32} Aside from viscoelasticity, complexities in

soft wetting arise when the elastomer is infused with a liquid, where phase separation is possible.^{33–35} In such a case, which is common for elastomers with moduli of ~ 100 kPa or less, both poroelastic and viscoelastic responses affect the wetting ridge dynamics.^{36–39} Although many efforts have been placed on the physics of soft wetting, the majority of experiments have been conducted with crosslinked elastomers or thin polymer sheets.^{40,41} On the other hand, studies on uncrosslinked, polymeric melts are limited, especially for wetting ridges larger than a few hundred nanometers at temperatures well above its glass transition temperature (T_g).^{42–44} Hence, the following questions remain: Can the linear rheology of a polymer melt help predict the growth rates of a wetting ridge? How do the viscoelastic properties of the polymer melt describe the shape of the wetting ridge profile?

Here we investigate how an immiscible liquid drop wets an amorphous polymer melt above T_g . We measure the shape of the wetting ridge profiles as a function of both the time of deposition (t_d) and the surface temperature (T). We show that the vertical growth dynamics of the wetting ridge are associated with the polymer dynamics, as characterized by shear rheology. Additionally, our results illustrate that the storage modulus (G'), measured at the relevant time scale, can be implemented to describe the overall shape of the wetting ridge profile.

Results and discussion

Experimental approach

In our experiments, we use poly(n-butyl methacrylate) (PBMA) as our model polymer surface. The polymer is first cast as a film with a thickness of ~0.9 mm. This thickness is chosen to minimize effects that the underlying glass substrate may have on the wetting behavior. PBMA is relatively stiff at room

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Fig. 1 (a) Glycerol droplet on a molten PBMA substrate at 125 °C at 5 minutes after deposition, just prior to removal from heat and blotting. (b) Top view of the indent left behind by the drop as seen under an optical microscope. (c) Profile of the indent by stylus profilometry, taken along the dotted line in (b). (d) Schematic showing the forces acting upon a wetting ridge near the three-phase contact line and the associated geometries. Scale bars in (a and b) are 500 μ m.

temperature with a $T_{\rm g}$ of ~28 °C, obtained by differential scanning calorimetry (Fig. S1, ESI⁺), and becomes softer as the temperature is raised. To conduct wetting studies, the substrate is placed on a hot plate and held at a desired temperature T for 2 minutes, which is sufficient for the film to reach an equilibrium temperature; a $\sim 4 \ \mu L$ drop of glycerol (which is also preheated to T) is then deposited onto the surface. Glycerol is used for its slower evaporation rate but similar surface tension to water, and because it does not swell the PBMA. To investigate multiple temperatures above the T_{g} , we use T = 65, 95 and 125 °C. These temperatures were chosen for the purpose of exploring a large temperature range while maintaining experimental feasibility. The three temperatures are above the glass transition temperature, hence there are no concerns of phase changes or glassy-to-rubbery transitions. After a certain deposition time, the substrate with the drop is removed from heat (Fig. 1a) to "freeze" the shape of the wetting ridge; the drop is then blotted off, leaving a circular footprint of an indent on the surface that can be visualized with standard optical microscopy (Fig. 1b). The circumference of the indent has an elevated wetting ridge, which is quantified using a stylus profilometer (Fig. 1c). Although optical systems like confocal microscopy and profilometry⁴⁵ have been used to measure wetting ridge profiles, most of the ridges in our case are too high and steep, making it difficult to capture the actual profile near the three-phase contact line. Additionally, many fluorescent dyes are not stable at higher temperatures. While atomic force microscopy (AFM) can offer higher resolution near a tip, both the lateral area and vertical heights are limited to sizes that are outside the range of our experiments. Therefore, optical profilometry is only used for wetting ridges that are less than 0.25 μ m in height, whereas the stylus is used for all other measurements. A comparison and discussion of the different methods considered for our samples is given in ESI† (Fig. S2). In general, it is inherently challenging to image the wetting ridge buried below a contact line;^{36,46} the ability to freeze it in place allows for measuring it directly.

Wetting ridge measurements

The formation of a wetting ridge is driven by the surface tension γ pulling up at the TPCL (Fig. 1d), which is balanced by the mechanical response of the polymer substrate. The ridge profile is described as h(x), where x is a position along the bottom axis. h_0 is the peak height occurring at the TPCL and is defined as the origin (Fig. 1d). This peak height is defined as the maximum positive deviation of the polymer surface from its initial state before the drop is deposited. To consider how a ridge height grows over the deposition time (t_d) as a function of T, the h(x) profiles are measured for different t_d at T = 65, 95and 125 °C. An example for the 95 °C wetting ridges is presented as zoomed-in images in Fig. 2a. The ridge height clearly increases with increasing t_d . For example, after a few minutes, h_0 is <1 μ m. After 20 min, $h_0 \sim 5 \mu$ m and reaches close to $\sim 20 \ \mu m$ after 3 hours. Similar trends are observed for wetting ridges for both the experiments at 65 °C and 125 °C, although with quantitatively different values for h_0 (Fig. S3, ESI[†]).



Fig. 2 (a) Growth of PBMA wetting ridges over time at 95 °C. The profiles have been plotted such that the *y*-axes for each line is zeroed at the *x*-axis point where the polymer film does not deviate from its initial height. Inset: The black lines denote logarithmic fits and the blue square is the intersection, which is taken as the peak height. This is an example after 180 minutes at T = 95 °C. (b) Peak heights of the ridges with increasing time for 3 different temperatures. Error bars correspond to standard deviation over three data sets.

An important point to note here is that the wetting ridges appear to have 'blunt' peaks when highly zooming into the contact line (Inset, Fig. 2a). However, this is unlikely the true tip because in theory, we expect the TPCL to have a negligible thickness on the order of a few nanometers. Hence, we expect that the peak heights are actually higher than those experimentally measured. This discrepancy arises due to limitations of the stylus profilometer where very small features become obfuscated, like the wetting ridge tip. To determine a more realistic peak, we fit a simple logarithmic function in the form $h = a + b \log(\pm(x - c))$ for both sides of the contact line (inset, Fig. 2a).⁴⁷ These fits extend beyond the experimental stylus peak in the vertical direction and their intersection provides a location of the expected ridge peak. A similar approach has been employed by van Gorcum et al.¹⁸ An example of the peak being located is shown in the inset of Fig. 2a for a 180 min sample at T = 95 °C, by zooming in into the top of the wetting ridge. While the maximum height of the wetting ridge from the experimental data is ~19 μ m, the peak found using logarithmic function fitting is ~24 μ m. A discussion of the calculated peak heights, the validity of this method, and a comparison of the experimentally measured peaks are provided in the ESI† (Fig. S4). For the remainder of this paper, the term 'peak' and the variable h_0 will refer to those determined by logarithmic extrapolation.

To gain more insight on the ridge growth dynamics, h_0 is plotted as a function of t_d for $T = 65^\circ$, 95° , and 125° C (Fig. 2b). As illustrated by the data in Fig. 2a and Fig. S3 (ESI^{\dagger}), h_0 continuously grows over time. For all tested temperatures, the ridge growth is initially fast, followed by a slower growth regime. In addition, the fast initial ridge formation appears to be strongly related to T. For example, after about 10 minutes, h_0 for the three temperatures are more than an order of magnitude different from each other (Fig. 2b): $h_0 \approx 0.2, 3,$ and 40 μ m for $T = 65^{\circ}$, 95°, and 125 °C, respectively. To test whether the ridge height reaches a plateau at long times, a 65 °C sample is held at elevated temperature for two days with the drop. Even after 48 hours, the ridge continues to grow monotonously, albeit at a slower rate (Fig. S5, ESI⁺). Hence, we expect that the ridge will continue to grow over time and wrap the drop, given a sufficiently long time. Nevertheless, the results in Fig. 2b suggest that there are different ridge growth dynamics at short and long times for the different temperatures, which are likely attributed to the polymer dynamics.

Relating ridge growth to polymer dynamics and viscoelasticity

When a liquid drop is deposited on a polymer melt, a finite force is applied instantaneously at the contact line at an initial time $t_{\rm d} = 0$. This force remains roughly constant after spreading, although the directionality may change with the contact angle. This constant force results in a time-dependent growth of the wetting ridge, analogous to a creep experiment. Hence, the ridge growth dynamics, defined by $h_0(t)$, will manifest in the viscoelastic properties of the polymer. The viscoelastic response of a polymer melt is defined by the frequency (ω)-dependent storage ($G'(\omega)$) and loss modulus ($G''(\omega)$). Based on the characteristics of the rheological data, we identify regimes of the polymer chain dynamics, and consequently relate them to the growth rates of the wetting ridge. For polymer melts in these temperatures, the likely chain relaxation regimes are Rouse and reptation modes,48 demarcated by the points of intersection (crossovers) of the storage and loss modulus. We use small amplitude oscillatory shear rheology on the PBMA to experimentally characterize the crossover points of G' and G''. To relate macroscopic rheological measurements to the polymer dynamics, we first consider the temperature of 95 °C. The rheology shows that low and high frequency crossovers occur at $\omega_{\rm c}^{\rm low}$ = 1.6 × 10⁻³ rad s⁻¹ and $\omega_{\rm c}^{\rm high}$ = 76 rad s⁻¹, respectively (Fig. 3a). Since our wetting experiments are conducted over time, these frequencies are inverted into the time-domain by taking $t = 1/\omega$. Hence, the crossover times for PBMA at 95 °C are $t_{\rm c}^{\rm low} \approx 625 \ {\rm s} \approx 10 \ {\rm min} \ {\rm and} \ t_{\rm c}^{\rm high} \approx 0.01 \ {\rm s}, \ {\rm where} \ t_{\rm c}^{\rm low} {\rm corresponds}$ to a reptation time $\tau_{\rm rep}$ and $t_{\rm c}^{\rm high}$ corresponds to a Rouse time $\tau_{\rm R}$.⁴⁸ These times imply that polymer chains undergoing deformation,



Fig. 3 (a) Dynamic oscillatory shear rheology of PBMA at 95 °C, with modulus crossover points marked by lines where G' = G''. (b) Peak wetting ridge heights *vs.* wetting deposition time for the three different temperatures. Lines denote scaling laws based on Rouse (dashed lines) and reptation (solid lines) regimes.

occurring between these timescales, will relax using Rouse modes. The viscoelastic response is due to the relaxation of chain segments, where the polymer chain diffusion coefficient scales with time as $D(t) \sim D_{\text{Rouse}} \sim t^{-1/2}$. For timescales longer than ~10 min, the chains will relax in the reptation regime with a constant self-diffusion coefficient $D(t) \sim D_c$. In this domain, the viscoelastic response is controlled by the chain diffusing on length scales of the order of its own size. The deformation of the polymer film h(t) can be considered to scale as the mean square displacement (MSD) of the polymer chains, which is proportional to the time elapsed:⁴³ $h_0(t) \sim \text{MSD} \sim D(t)t$. Thus, in the Rouse limit, $h_0(\tau_R) \sim D_{\text{Rouse}}(t)t \sim t^{1/2}$. Beyond Rouse timescales, reptation modes dominate and the ridge is predicted to grow as $h_0(\tau_{\text{rep}}) \sim D_c t \sim t$.

Scaling law fits of the peak height data h_0 at 95 °C, presented in Fig. 3b, shows that for the first ~ 600 s the peak height scales as the square root of time, $h_0 \sim t^{1/2}$, consistent with Rouse behavior. For time scales beyond that, the scaling law changes to $h_0 \sim t$, which is indicative of reptation. This is in agreement with the predictions from the crossover points of rheological experiments. Hence, we confirm there are two regimes of the ridge growth rate. The effect of temperature on this demarcation between regimes is realized from the rheology on PBMA at lower (65 °C) and higher (125 °C) temperatures (Fig. S6, ESI†). At 65 °C, a low frequency regime is not observed even at frequencies as low as 5×10^{-4} rad s⁻¹, indicating the absence of a reptation regime within the timescale of our corresponding wetting experiment. Hence the growth of all peaks at 65 °C scale as $h_0 \sim t^{1/2}$ (Fig. 3b). Conversely, for 125 °C, the absence of a high frequency crossover suggests a singular reptation-dominated growth rate scaling of $h_0 \sim t$ (Fig. 3b).

For the 125 °C sample, the experimental wetting ridge data clearly deviates from the $h_0 \sim t$ scaling at long times (Fig. 3b). We attribute this to the high testing temperature; at 125 °C, glycerol drops evaporate after prolong testing. Based on literature, we estimate the rate of evaporation for glycerol at 125 °C as ~1900 g s⁻¹ m⁻²,⁴⁹ with a vapor pressure 10 000 times greater than at room temperature.⁵⁰ For times $t_d \gtrsim 15$ min, glycerol evaporation can be observed; the wetting ridge, now devoid of any pinned liquid, starts to relax by re-flowing back into the substrate.

Wetting ridge profile

At this stage, we consider how to describe the profile of the wetting ridge as the ridge grows over time. In the 1980s and 90s, Shanahan, de Gennes, and others developed an expression for the shape of a wetting ridge on a crosslinked rubber, 19,27,29,47 which has a constant long-time (low-frequency) modulus; this concept has since been further explored by several researchers.^{3,51,52} For example, Limat expanded this effort to mitigate issues of a singularity at the contact line, rather than implementing a cut-off length.⁵³ In these models, the profile scales as γ/G , where the shear modulus is taken as timeindependent, $G'(\omega \rightarrow 0)$. Due to the predominant solid-like behavior of a crosslinked rubber at long times, $G''(\omega)$ is usually orders of magnitude smaller than $G'(\omega)$. However, this is not the case for a polymer melt. Here we employ the storage modulus; however, instead of taking a single value at $\omega \rightarrow 0$, we use the time-dependent value $G'(\omega_d = 1/t_d)$, where t_d are the discrete time steps in our wetting experiment. We expect that the corresponding values of G' measured by rheology will reflect the time-dependent response during wetting. Hence, we propose that $G'(1/t_d)$ can be implemented as a simple approach to capture the ridge profile. To test this hypothesis, we insert G' into the model from Limat to describe the shape of the air-polymer wetting ridge. Specifically, the profile h(x) at a given time and temperature is then given by:

$$h(x, t_{\rm d}) = \frac{\gamma_{\rm LV} \sin \theta}{2\pi G'(1/t_{\rm d})} \ln \left(\frac{d+l_{\rm s}}{|x|+l_{\rm s}}\right) \tag{1}$$

where γ_{LV} is the surface tension of glycerol in air at the given temperature, *d* is the positive *x*-distance at which the substrate is flat, and l_s is the elastocapillary length⁵³ defined by $l_s = \gamma_{SV}/(\pi G')$, where γ_{SV} is the surface tension of the PBMA. To predict the profile from eqn (1), γ_{LV} and γ_{SV} are taken from

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Fig. 4 Ridge profiles (black data points) near the TPCL obtained at the three temperatures for different wetting times: (a) $65 \degree$ C, (b) $95 \degree$ C, and (c) $125 \degree$ C. Dotted lines (red) are theoretical fits using eqn (1) on the polymer-air side of the TPCL. Triangles (blue) indicate the predicted peak height from logarithmic fitting. Data sets are shifted in the *y*-axes for clarity.

literature, noting that the surface tensions are temperature dependent. Extrapolating from the measured values by Wu,^{54,55} we calculate γ_{SV} values for PBMA to be 28.5, 26.8, 25.0 mN m⁻¹ for $T = 65^{\circ}$, 95°, and 125 °C, respectively. The liquid surface tension values for glycerol vary as 60.0, 57.2, 54.1 mN m⁻¹ for the three temperatures, respectively.⁵⁶ The values for θ , $G'(1/t_d)$, and d are obtained directly from our experiments. To examine how eqn (1) compares to experiments, we plot the experimental and calculated wetting profiles for selected deposition times for all temperatures in Fig. 4, noting here that there are no fitting parameters; all variables are either measured directly or determined from literature. Eqn (1) overlays the experiments reasonably well, illustrating that h(x) can be described by incorporating G' values, at the appropriate temperature and time scale, into a model typically used for elastic solids. The model agrees with the experimental data for most of the profile; however, there are clear deviations in the peak prediction. It can be seen that the maximum heights of the wetting ridge (h(x = 0)) predicted by eqn (1) is often lower than the peak heights estimated using the logarithmic fitting (blue triangles), which may be considered as the "true" theoretical peaks. This discrepancy may arise due to the choice of the correction factor - the elastocapillary length. This length scale can be defined as $l_s^{el} = Y/G$ for elastic solids,⁵⁷ where Y is the surface stress. Y has been reported to be strain-dependent for elastic solids, although with some discrepancies.46,58-60 We employ an analogous elastocapillary length for polymer melts in eqn (1), using the solid-vapor surface tension γ_{sv} and storage

modulus G'. Using this surface tension does not take into account any strain-dependencies that may exist in a potentially non-relaxed state. Additionally, the modulus may not be spatially homogeneous throughout the wetting ridge, which would alter the predicted height; however, this is difficult to verify. From an experimental point of view, a potential source of error arises from small variations in the measured G' from rheology. Moreover, while care has been taken during substrate preparation, tilts of the order of $\sim 0.1^{\circ}$ are observed on the surface. These compounded errors may result in the experimental data departing from the eqn (1) predictions. Nevertheless, we find that the experimental profiles agree rather well with eqn (1). While there are more exact frameworks^{53,61,62} to describe the shape of the wetting ridge, we show here that incorporating the time-dependent modulus provides a simple and direct way to relate how the rheology of a polymer melt governs its wetting behavior.

Conclusions

We measure the growth dynamics of a wetting ridge caused by the deposition of a liquid drop on an uncrosslinked polymer melt at temperatures well above its glass transition. The wetting ridges grow over time and seem to grow continuously, without reaching any apparent equilibrium height. Using scaling laws associated with polymer dynamics, two regimes of ridge growth rate exist within a single temperature. These two regimes are associated with Rouse dynamics $(h \sim t^{1/2})$ and reptation $(h \sim t)$, as defined by crossover points in shear rheology. The storage modulus, measured at the appropriate timescale, is incorporated into a model to predict the shape of the wetting ridge profile. The model was reported by Shanahan and coworkers and expanded by Limat to describe wetting ridges of elastomers. Our modified equation fits the experimental data over multiple temperatures and timescales with reasonable agreement. The temperatures explored here are higher than those in prior studies, and closer to the temperatures used in polymer melt processing. We anticipate that our findings will aid in commercial applications which involve surface interactions of polymer melts, including blending, injection mold design, additive manufacturing, and high temperature coatings.

Conflicts of interest

There are no conflicts to declare.

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