

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this manuscript, experimentally the authors find a counter-intuitive phenomenon where a surrounding viscous phase can switch the overall speed of dewetting such that films retract slower as the surface repellency increases. The mechanism is revealed to be the different dissipation origins with different viscosity ratio. The topic is interesting, and could attract the attention of the readership of Communications Physics. It would be good if the author could address the following issues before being accepted.

1. Figure 2 shows the influence of viscosity and contact angle on the timescale of dewetting process. However, many other factors may influence the experiment results, such as temperature, surface topography or roughness, thickness deviation of the liquid film, liquid density and so on. It is better to estimate the influence of these factors and get the error bar of your experiment.
2. Surface features are important for the experiments, it is better to give some figures of surface characterization.
3. In the manuscript, the author presents a different tendency of relaxation time between two different M . Is there a critical M which separate this difference tendency which will be meaningful?

Reviewer #2 (Remarks to the Author):

The authors present experimental and theoretical results of studies of the dewetting dynamics of droplets/films surrounded by viscous liquids onto substrates of different wetting properties. As part of their results they observe an unexpected effect in which the dewetting in the non-wetting case is slowed down (compared to the wetting case) by the viscous (surrounding) fluid due to the geometry of flow near and around the contact line.

The article is interesting and good agreement between experiments and simple model are presented - however, the result while surprising is not sufficiently novel. I believe these results be better suited for publication in a dedicated journal such as Physics of Fluids.

Specific comments:

- 1- A sketch pinpointing the variables used in Eq1, and Eq3 would be very valuable. How valid is the small angle approximation here? Based on the images it is not obvious this is easily applicable.
- 2- Has the error of the experimental measurements of the relaxation time due to the fitting being considered? In other words, where do the error bars in Fig 2 come from (they are the same always, so this must come from somewhere else?).

Reviewer #3 (Remarks to the Author):

The paper describes a unique dewetting behaviour when a liquid droplet is surrounded by another viscous liquid. The results and the content of the paper is relevant to the journal. While the paper is fit for publication, I have a few questions to augment the discussion:

- 1) How does the curvature of the interface (during the retraction at the contact line) change with time? Is there a scaling law?
- 2) In fig 1a, at time $t=0$ the liquid-liquid interface close to the contact line has a curvature such that there appears to be the surrounding viscous phase below the droplet edge. Does the retraction result in the ingress of the surrounding fluid?

We would like to thank the Reviewers for their careful reading and constructive feedback on our paper. Below we provide a point by point response to all reviewers' comments. Changes to the manuscript are highlighted in blue.

Reviewer #1 (Remarks to the Author):

In this manuscript, experimentally the authors find a counter-intuitive phenomenon where a surrounding viscous phase can switch the overall speed of dewetting such that films retract slower as the surface repellency increases. The mechanism is revealed to be the different dissipation origins with different viscosity ratio. The topic is interesting, and could attract the attention of the readership of Communications Physics. It would be good if the author could address the following issues before being accepted.

1. Figure 2 shows the influence of viscosity and contact angle on the timescale of dewetting process. However, many other factors may influence the experiment results, such as temperature, surface topography or roughness, thickness deviation of the liquid film, liquid density and so on. It is better to estimate the influence of these factors and get the error bar of your experiment.

The main effect that can influence the experimental results comes from variation in density, surface tension and viscosity as a consequence of changes in temperature. The experimental results reported in the paper were carried out at a temperature $T = 21.0\text{ }^{\circ}\text{C} \pm 0.5\text{ }^{\circ}\text{C}$.

We measured the variation in density of TMP-TG-E with temperature using an Anton Parr DMA 4500 density meter over the range 10°C to 25°C . We find that the density of TMP-TG-E varies linearly with temperature, resulting in a variation in density of 0.00043 g/cm^3 per $0.5\text{ }^{\circ}\text{C}$ in our experiments. The density of PDMS oils was obtained from the manufacturer (safety datasheets; Alfa Aesar) where the density values are stated to be 0.97 g/cm^3 at 20°C . The variation of the density of the PDMS oils with temperature can be found in Ref [1], where again a linear relationship between temperature and density is found for a variety of PDMS oil viscosities, again with a small variation of 0.00046 g/cm^3 per 0.5°C . However, because gravitational and inertial effects are negligible in our experiments due to the small droplet sizes and the dominant effect of viscosity and surface tension, the small variations in density do not significantly contribute to the error of our measurements.

To quantify the effect of variations in the viscosity of the liquids, we measured the viscosity of all PDMS oils and TMP-TG-E between 20°C and 22°C using a TA Instruments CSL² 100 rheometer. We find that a 0.5°C temperature difference about 21°C leads to a maximum possible error of 4% on the value of TMP-TG-E viscosity (μ_i), on average a 1% error on the value of μ_o . Standard propagation of these errors leads to a maximum error in the viscosity contrast, $M = \mu_o/\mu_i$, of 5%. This error is smaller than of the size of the symbol reported Fig. 3.

The variation of the surface tension of TMP-TG-E with temperature was measured using a DuNoüy ring tensiometer (Krüss K6) combined with a temperature-controlled bath over the range 0°C to 37.7°C . We find that the surface tension of TMP-TG-E in air varies linearly by $0.1\text{ mN/m}^{\circ}\text{C}$. We measured the interfacial tension between TMP-TG-E and PDMS using the pendant droplet technique (Krüss DSA10-MK2) at a temperature of 21°C . We took 40 separate measurements of the interfacial tension in two different viscosity grades of PDMS finding that the interfacial tension did not vary between PDMS viscosity grades and that the interfacial tension is $6.2 \pm 0.3\text{ mN/m}$. While temperature may influence the interfacial tension between the two fluids, we expect this effect to be linear and of the order of 0.1 mN/m per degree Celsius. As the measurement of the interfacial tension (0.3 mN/m) is the larger error, we consider this as the main source of error in our measurements.

Propagating the uncertainty in the viscosity of the inner liquid and the interfacial tension leads to an uncertainty in the capillary number of 9%, which is smaller than the size of the symbol in Figure 3a.

We have added this discussion of the uncertainty of the parameters in our experiments to the methodology, and a note to the figure explaining the error bars.

2. Surface features are important for the experiments, it is better to give some figures of surface characterization.

The surface roughness has been measured in three areas for each sample using a Dektak surface profilometer scanned over a 6 mm area with a horizontal step of 0.16 μm . These measurements show that at small scales (over nanometres) the surface has a mean deviation surface roughness (R_a) of 39.5 ± 0.8 nm for the NOVEC 2702 surface and 26 ± 9 nm for the Teflon AF surface. Therefore, at the small scale the two surfaces have a similar roughness.

At larger scales the Dektak surface measurements show larger features, of the order of microns. These are not due to the electrode structure, nor due to the SU-8 dielectric (capping and planarization) layer. Instead these features appear from the drying of the final top surface coatings, NOVEC and Teflon. We have quantified how these features can locally pin the contact line momentarily during small movements, contributing to small scale noise in the measurement of the instantaneous film radius close to equilibrium. To account for this effect, we take the measurement of the relaxation time in a range above the final relaxation ($0.01 < R(t) - R_e < 0.2$) as shown in Fig. 1, which avoids any systematic effects from these features in the large-scale film dynamics.

We have added a discussion about the surface characterisation to the Materials and Methods section of the manuscript.

3. In the manuscript, the author presents a different tendency of relaxation time between two different M. Is there a critical M which separate this difference tendency which will be meaningful?

As the reviewer points out, the relaxation time shows two distinct asymptotic behaviours (at low and large M); however, we found that the cross over between both regimes is gradual and varies depending on the wettability of the surface (compare the data on Teflon and NOVEC surfaces, respectively). Hence, we cannot identify a single critical value of the viscosity ratio separating both regimes. We have added a sentence to the manuscript (last sentence in the paragraph that follows eq (8)) to clarify this point.

[1] Roberts, C.; Graham, A.; Nemer, M.; Phinney, L.; Garcia, R.; Stirrup, E. Physical Properties of Low-Molecular Weight Polydimethylsiloxane Fluids. Technol. Rep. 2017 <https://doi.org/10.2172/1343365>

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The authors present experimental and theoretical results of studies of the dewetting dynamics of droplets/films surrounded by viscous liquids onto substrates of different wetting properties. As part of their results they observe an unexpected effect in which the dewetting in the non-wetting case is slowed down (compared to the wetting case) by the viscous (surrounding) fluid due to the geometry of flow near and around the contact line.

The article is interesting and good agreement between experiments and simple model are presented - however, the result while surprising is not sufficiently novel. I believe these results be better suited for publication in a dedicated journal such as Physics of Fluids.

Specific comments:

1- A sketch pinpointing the variables used in Eq1, and Eq3 would be very valuable. How valid is the small angle approximation here? Based on the images it is not obvious this is easily applicable.

We have produced a sketch explaining the variables in Eqs 1 and 3 (new figure 2).

The small angle approximation is presented to supplement the quantitative agreement shown by the full equations (4) and (8), which we plot in Fig. 3, by illustrating the change in behaviour of the interface speed and relaxation time with viscosity ratio. The reviewer is correct to point out that these simplified expressions would not hold to describe our whole set of experimental data (as the angle varies from 56° to 137°) and we now point this out in the paper.

2- Has the error of the experimental measurements of the relaxation time due to the fitting being considered? In other words, where do the error bars in Fig 2 come from (they are the same always, so this must come from somewhere else?).

The error in the fitting of the exponential relaxation has been considered and the error bars shown in Fig 2b come from the error in the fitting. The fitting procedure is as follows, we first plot a graph of $\ln(R(t) - R_e)$ against time, where we first use a trial value of R_e which is the final measured value of R_e . We then fit a straight line to between $(0.01 < R(t) - R_e < 0.2)$. We then change the value of R_e until the correlation coefficient (R^2) is maximised which gives the value of the relaxation time, τ .

We then take an error margin of δR_e about the determined value of R_e , carrying out the fitting and computing the relaxation time from both $R_e - \delta R_e$ and $R_e + \delta R_e$. These fits then give the uncertainty in the relaxation time, $\delta\tau$. This approach naturally results in large errors of the value of τ which are on average 20% of the calculated value of τ .

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2) In fig 1a, at time $t=0$ the liquid-liquid interface close to the contact line has a curvature such that

there appears to be the surrounding viscous phase below the droplet edge. Does the retraction result in the ingress of the surrounding fluid?

Response to 1&2:

The curvature of the contact line (in the plane of the solid surface) increases with time due to the retraction of the film. At the same time, the transverse curvature of the rim (in a plane parallel to the radial coordinate) decreases as the rim increases. Here we have focused on the effect of the viscosity ratio on the overall dewetting dynamics, and thus leave the details of the scaling of the curvature as an open question.

There is no outer liquid trapped underneath the film as the inner liquid film (TMP-TG-E) is first slowly spread in air e.g. starting from 0V and increasing quasistatically until fully spread at V_{th} . This fully spread film is then slowly immersed in the surrounding PDMS liquid. As TMP-TG-E is more polarisable than the PDMS, TMP-TG-E remains fully spread during and after the submersion step.

We have included a note detailing this experimental step to prevent the entrapment of the outer liquid in the manuscript.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed the issues I raised. I would like to recommend it for publication. One suggestion is that it would be good to add a short discussion about the influences of variation in density, surface tension and viscosity as a consequence of changes in temperature on the experimental results as stated in the authors' reply to my first question.

Reviewer #2 (Remarks to the Author):

while the authors have answered my concerns satisfactorily (although I would suggest they describe their choice of computing errors in τ in their paper, perhaps as supplemental material?) if not in the main body of the paper. However, after having read the manuscript in detail again I still suggest that the results would be better suited in a fluid-oriented journal.