On the interaction of viscoelasticity and waviness in enhancing the pull-off force in sphere/flat contacts

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Abstract
Motivated by roughness-induced adhesion enhancement (toughening and strengthening) in low modulus materials, we study the detachment of a sphere from a substrate in the presence of both viscoelastic dissipation at the contact edge, and roughness in the form of a single axisymmetric waviness. We show that the roughness-induced enhancement found by Guduru and coworkers for the elastic case (i.e. at very small detachment speeds) tends to disappear with increasing speeds, where the viscoelastic effect dominates and the problem approaches that of a smooth sphere. This is in qualitative agreement with the original experiments of Guduru’s group with gelatin. The cross-over velocity is where the two separate effects are comparable. Viscoelasticity effectively damps roughness-induced elastic instabilities, and make their effects much less important.

Keywords:
Roughness, Adhesion, Guduru’s theory, viscoelasticity

1. Introduction

It is well known that adhesion of hard solids is difficult to measure at macroscopic scales, and Fuller and Tabor (1975) proved that even in low modulus materials (they used rubbers with $E \sim 1 \text{ MPa}$), a $\sim 1[\mu m]$ of roughness destroy adhesion almost completely, despite van der Waals adhesive forces are quite strong, giving the so called ”adhesion paradox” (Kendall, 1975). Adhesion of macroscopic bulk objects requires smooth surfaces, and at least one of the solids has to have a very low elastic modulus. Dahlquist
(1969a, 1969b) while working at 3M proposed a criterion largely used in the
world of adhesives, namely that the elastic Young modulus should be smaller
than $\sim 1 \text{ MPa}$ to achieve stickiness even in the presence of roughness. This
clearly is just a rough indication, but Tiwari et al. (2017) find for example
that the work of adhesion (at a given retraction speed) is reduced of a fac-
tor 700 for a rubber in contact with a rough hard sphere when the rubber
modulus is $E = 2.3 \text{ MPa}$, but is actually increased because of roughness
by a factor 2 when the rubber has modulus $E = 0.02 \text{ MPa}$. The threshold
doesn’t change much even if we consider nanometer scale roughness, as in the
recent results of Dalvi et al. (2019) for pull-off of PDMS hemispheres having
four different elastic moduli against different roughened plates: Dahlquist’s
criterion seems to work surprisingly well, as while there is little effect of
roughness for the 3 cases of low modulus up to near $E = 2 \text{ [MPa]}$, roughness
has strong effect both during approach and retraction for the high modu-
lus material ($E = 10 \text{ [MPa]}$), where the hysteresis may be due partly to
viscoelastic effects. However, for the 3 low modulus materials, roughness
almost systematically increases the work of adhesion rather than decreasing
it as for the high modulus material, for a given retraction speed.

Roughness-induced adhesion enhancement was measured with some sur-
prise first by Briggs & Briscoe (1977), and Fuller & Roberts (1981), and
Persson-Tosatti’s (2001) theory above attributes it to the increase of sur-
face area induced by roughness. Another mechanism was put forward by
Guduru and collaborators (Guduru, 2007, Guduru and Bull, 2007). Guduru
considered a spherical contact having a concentric axisymmetric waviness,
and considers the contact is complete over the contact area. The waviness
gives rise to oscillations in the load-approach curve which result in up to
factor 20 increase of the pull-off with respect to the standard smooth sphere
case of the JKR theory (Johnson, Kendall & Roberts, 1971). Also, the curves

\footnote{Despite the authors intended to remove as much as possible rate-dependent effects
by applying a retraction rate of only $60 \text{ nm/s}$). The authors claim a good correlation
of the energy loss during the cycle of loading and withdrawing with the product of the
real contact area at maximum preload with the "intrinsic" work of adhesion. Notice that
this would not work for a smooth sphere where JKR theory predicts that the energy loss
is independent on preload and indeed the data of Dalvi et al. (2019) with the lowest
roughness do show almost a constant trend. Also, the hard material case shows almost no
energy loss.}

\footnote{Whereas adhesion reduction is attributed by Persson and Tosatti (2001) to the elastic
energy to flatten roughness, which is proportional to the elastic modulus.}
fold on each other so that we expect jumps at some points in the equilibrium curve, which corresponds to dissipation and emission of elastic waves in the material, and results in strong hysteresis. Later, Kesari and Lew (2011) noticed that Guduru’s solution has an elegant “envelope” obtained by expanding asymptotically for very small wavelength of the waviness.

But most soft materials are viscoelastic, and therefore there is a strong velocity dependence of the pull-off result. Many authors (Gent and Schultz, 1972, Barquins and Maugis 1981, Gent, 1996, Gent & Petrich 1969, Andrews & Kinloch, 1974, Barber et al, 1989, Greenwood & Johnson, 1981, Maugis & Barquins, 1980, Persson & Brener, 2005) have proposed that the process of peeling involves an effective work of adhesion \( w \) which is the product of the thermodynamic (Dupré) work of adhesion \( w_0 \) and a function of velocity of peeling of the contact/crack line and temperature, as long as there is no bulk viscoelasticity involved, over a large range of crack speeds, namely of the form that has been validated also by a large amount of data including peeling tests at various peel angles

\[
w = w_0 \left[ 1 + k (a_T v_p)^n \right]
\]

where \( k, n \) are constants of the material, with \( n \) in the range \( 0.1 - 0.8 \) and \( a_T \) is the WLF factor (Williams, Landel & Ferry, 1955) which permits to translate results at various temperatures \( T \) from measurement at a certain standard temperature. This form of effective work of adhesion was obtained also from theoretical models using either Barenblatt models or crack tip blunting models (Barber Donley and Langer 1989, Greenwood and Johnson, 1981, Persson & Brener, 2005) generally using simple a single relaxation time model for the materials. Actually, Persson & Brener (2005) showed that for a frequency dependent viscoelastic modulus \( E(\omega) \sim \omega^{1-s}, \ 0 < s < 1 \), in the transition region between the ”rubbery region” and the ”glassy region” (where the strong internal damping occurs important for energy loss processes), the equation (1) is satisfied at intermediate velocities with \( n = (1-s)/(2-s) \) (so that \( 0 < n < 1/2 \), in agreement with most of the range cited above). There may be deviations for materials having a complex behaviour with many relaxation times or for considering the role of finite size of the system or of very high temperature at the crack tip which depends on thermal diffusivity of the material, see recent review (Rodriguez et al., 2020).

The effective “toughness” \( w \) can increase of various orders of magnitude over \( w_0 \) as the velocity increases (more precisely, of the ratio \( E(\infty)/E(0) \),
where \( E(\omega) \) is the frequency dependent elastic modulus), and the pull-off of a sphere has also been effectively measured to increase of various orders of magnitude over an increase of peeling speed (Barquins & Maugis, 1981). On the contrary, during crack closure the effective work of adhesion is even smaller than \( w_0 \), (this time it is reduced by the ratio \( E(0)/E(\infty) \), see Greenwood and Johnson, 1981), so in some cases loading could become essentially an elastic model without adhesion.

Equation (1) generalizes the thermodynamic equilibrium of elastic cracks for the strain energy release \( G \): namely, it provides a condition for crack edge velocity — when \( G > w_0 \), the crack accelerate under the force \( G - w_0 \) applied per unit length of crack, until a limit speed \( v_p \) for equilibrium is found, depending on the loading conditions. For example, \( G - w_0 \) is a constant for classical peeling experiments, whereas it monotonically increases for flat punches, and has a much richer behaviour for the smooth sphere. Therefore, for imposed tensile load smaller in absolute value than the JKR pull-off value \( P_0 = 3/2\pi wR \), the contact area simply decreases to another equilibrium value (given asymptotically by JKR theory), while for imposed load below the JKR value, it decreases with non monotonic velocity but without the JKR pull-off instability, so up to complete detachment. Therefore, pull-off depends on the loading condition: can be anything greater than \( P_0 \) if load is imposed, whereas it is a precise function of the retraction rate in an experiment where the cross-head of a rigid machine keeps the remote approach velocity as constant.

Various authors (Barquins & Maugis, 1981, Greenwood & Johnson, 1981, Muller, 1999) have studied the peeling of viscoelastic spheres with the above form of fracture mechanics formulation (1), and some approximate scaling results have also been given (Muller, 1999), but a theoretical or numerical investigation about the coupled effect of viscoelasticity and roughness has not been attempted, in the best of the author’s knowledge, not even with numerical simulations. It seems that in general viscoelasticity can only increase the ”tack” i.e. the force or the work needed to detach two solids, whereas the role of roughness is more controversial, as we have discussed above. We are aware of the complexity of the general problem, so here, we tackle the study of a simple problem, that of a sphere with a single wave of roughness, which generalizes the relatively recent work of Guduru and collaborators (Guduru, 2007, Guduru and Bull, 2007) and following related literature, to the case of a viscoelastic substrate.
2. The theory

We consider the Guduru contact problem for a sphere against a flat surface, where the gap is defined as $f(r) = \frac{r^2}{2R} + A \left(1 - \cos \frac{2\pi r}{\lambda}\right)$, where $R$ is the sphere radius, $\lambda$ is wavelength of roughness, $A$ is its amplitude.

![Diagram of the problem](image)

Fig.1 - The geometry of the problem. A sphere of radius $R$ with a simple roughness being a single axisymmetric wave with wavelength $\lambda$ and amplitude $A$.

The Guduru problem can be solved by considering the stress intensity factor $K$ at the contact edge (radius $r = a$) or equivalently the strain energy release rate $G$ (Guduru, 2007)

$$G(a, P) = \frac{K(a, P)^2}{2E^*} = \frac{(P_1(a) - P)^2}{8\pi E^*a^3}$$

(2)

where $E^* = E/(1 - \nu^2)$ is plane strain elastic modulus (i.e. $E$ is Young’s modulus and $\nu = 0.5$ Poisson ratio generally equal to 0.5 in rubbery materials, while we consider the countersurface is generally much more rigid so we neglect its elastic properties).

Here, $P_1(a)$ is the load required to maintain a contact radius $a$ in the absence of adhesion, while $P$ is the smaller load to maintain the same contact
radius in the presence of adhesion. In particular standard contact mechanics gives (Guduru, 2007)

\[ P_1 (a) = 2E^* \left\{ \left( \frac{2}{R} + \frac{4\pi^2 A}{\lambda^2} \right) \frac{a^3}{3} + \frac{\pi A}{2} aH_1 \left( \frac{2\pi a}{\lambda} \right) - \frac{\pi^2 A a^2}{\lambda} H_2 \left( \frac{2\pi a}{\lambda} \right) \right\} \]  

(3)

where \( H_n \) are Struve functions of order \( n \).

In the adhesionless conditions, the remote approach (positive for compression) is

\[ \alpha_1 (a) = \frac{a^2}{R} + \frac{\pi^2 A}{\lambda} aH_0 \left( \frac{2\pi a}{\lambda} \right) \]  

(4)

so in the adhesive condition we have to decrease this by an amount given by a flat punch displacement giving the general result for approach

\[ \alpha (a, P) = \frac{a^2}{R} + \frac{\pi^2 A}{\lambda} aH_0 \left( \frac{2\pi a}{\lambda} \right) - \frac{P_1 (a) - P}{2E^* a} \]  

(5)

From (5), we can obtain the general equation for the load as a function of contact radius and approach

\[ P (a, \alpha) = P_1 (a) + 2E^* a\alpha (a) - 2E^* \frac{a^3}{R} - \frac{\pi^2 2E^* A}{\lambda} a^2 H_0 \left( \frac{2\pi a}{\lambda} \right) \]  

(6)

where \( P_1 (a) \) is given by (3) above. Imposing the condition of thermodynamic equilibrium \( G (a) = w_0 \) using (2) and (3) permits to write the Guduru solution explicitly as parametric equations of the contact radius \( a \)

\[ P (a) = P_1 (a) - a^{3/2} \sqrt{8\pi w_0 E^*} \]  

\[ \alpha (a) = \alpha_1 (a) - a^{1/2} \sqrt{2\pi w_0 / E^*} \]  

(7)

(8)

Using the Kesari & Lew (2011) expansion, Ciavarella (2016) obtained that the Guduru solution has oscillations bounded between two exact JKR (Johnson, Kendall & Roberts, 1971) envelope curves for the smooth sphere, but with a corrected (enhanced or reduced, respectively for unloading or loading) surface energy

\[ P_{env} (a) = \frac{4}{3R} E^* a^3 - a^{3/2} \sqrt{8\pi w E^*} \left( 1 \pm \frac{1}{\sqrt{\pi \alpha_{KLJ}}} \right) \]  

(9)

\[ \alpha_{env} (a) = \frac{a^2}{R} - a^{1/2} \sqrt{\frac{2\pi w}{E^*}} \left( 1 \pm \frac{1}{\sqrt{\pi \alpha_{KLJ}}} \right) \]  

(10)
where
\[
\alpha_{KLJ} = \sqrt{\frac{2w_0\lambda}{\pi^2 E^* A^2}}
\]  
(11)
is the parameter Johnson (1995) introduced for the JKR adhesion of a nominally flat contact having a single scale sinusoidal waviness of amplitude \( A \) and wavelength \( \lambda \). Thus, since (9,10) are JKR equation for a smooth sphere of radius \( R \), the factor
\[
\frac{w_{\text{eff}}}{w_0} = \left(1 + \frac{1}{\sqrt{\pi \alpha_{KLJ}}}ight)^2
\]  
(12)
is an roughness-induced increase which holds as long as a compact contact area can be obtained, which requires not too large roughness, and/or sufficiently strong precompression. In practice, factors up to 20 have been obtained also experimentally by Guduru & Bull (2007), although of course these were achieved in geometry built for the specific goal to achieve very large enhancement. Fig.2 elucidates the behaviour of the oscillations in the Guduru solution for a representative case, which we shall later extend to the viscoelastic solution. Given these gulfs and reentrances, in the elastic solution, the real followed path will depend on the loading condition. For a soft system (close to "load control"), there will be horizontal jumps in approach while in a stiff system (close to "displacement control") there will be vertical jumps to the next available stable position. In both cases there will be areas "neglected" during these jumps which represent mechanical dissipated energy. Indeed, in the "envelope" solution of Kesari-Lew-Ciavarella, the combined effect of these jumps results in the different JKR loading and unloading curves which give an additional hysteresis with respect to the standard JKR case, where the only hysteresis comes a single elastic instability in pull-in and another (different) single instability at pull-off. The dashed lines in Fig.2 are the Kesari-Lew-Ciavarella envelopes (9,10) using (11).
Fig. 2 - The load-approach curve in the Guduru elastic problem with $E^* = 16500 \text{ [Pa]}$; $R = 0.23 \text{ [m]}$; $w_0 = 0.008 \text{ [J/m}^2\text{]}$; $\lambda/R = 0.002$; $A/\lambda = 0.005$, and reference $a_i = 0.01 \text{ [m]}$.

2.1. Viscoelastic problem

For a given remote applied withdrawing of the sphere $v = -\frac{da}{dt}$, we can write the velocity of the contact edge as

$$v_p = -\frac{da}{dt} = v \frac{da}{d\alpha}$$ (13)

The condition $G(a) = w$ (which replaced the thermodynamic equilibrium $G(a) = w_0$ for the elastic sphere) therefore defines a differential equation for $a = a(\alpha)$, obtained using (2,1,13)

$$\frac{1}{k^{1/n}aTv} \left( \frac{P_1(a) - P_1^2}{8\pi E^*a^2w_0} - 1 \right)^{1/n} = \frac{da}{d\alpha} \quad (14)$$

Hence, using (6) and defining dimensionless parameters

$$V = k^{1/n}aTv \quad ; \quad \zeta = \left( \frac{2\pi w_0}{RE^*} \right)^{1/3} \quad (15)$$

we write (14) as

$$\frac{da}{d\alpha} = \frac{1}{V} \left[ \left( \frac{R/a}{\zeta^3} \left( \frac{\alpha}{R} - \frac{a^2}{\lambda R} - \pi^2 \frac{A}{\lambda R} H_0 \left( \frac{2\pi a}{\lambda} \right) \right)^2 - 1 \right]^{1/n} \quad (16)$$
which can be solved with a numerical method. After a solution is obtained for \( a = a(\alpha) \), we substitute back into (13) to compute the load. Notice that, for a given starting point of the peeling process in terms of load \( P \), the term under parenthesis in (14) is zero, and hence \( \frac{da}{d\alpha} \) starts off zero giving some delay with respect to the elastic curve, which is hard to eliminate even at very low withdrawal speeds.

3. Results

3.1. Rough sphere

We consider first a viscoelastic material having \( n = 0.33 \), and dimensionless withdrawal velocity \( V = 0.0002, 0.002, 0.02, 0.2, 2 \); the other constants, as indicated in Fig.1, are \( E^* = 16500 \text{[Pa]} \); \( R = 0.23 \text{[m]} \); \( w_0 = 0.008 \text{[J/m}^2] \); \( \lambda/R = 0.002 \); \( A/\lambda = 0.005 \). This corresponds to an “adiabatic” elastic enhancement of the pull off according to the equation derived from the Guduru theory (12) of \( w_{eff}/w_0 = 1.42 \). We indicate with \( P_0 = 3/2\pi w_0 R \) the JKR value of pull-off for the smooth sphere. The loading curve follows the elastic solution (7, 8), and we start withdrawing the indenter from a reference value of \( a_i = 0.01 \text{[m]} \). Corresponding values of initial approach \( \alpha_i \) and load \( P_i \) can therefore found from (7, 8). Numerical solutions are found with the NDSolve algorithm in Mathematica with default options. Fig.3a shows the obtained load-approach curve in terms of \( P/P_0 \) and \( \alpha/\alpha_i \) where \( P_0 \) is JKR pull-off of the smooth sphere, and Fig.3b shows the contact radius \( a/a_i \) peeling as a function of approach. The inner black wavy curves are the equilibrium Guduru solutions, and the other 5 curves are obtained numerically for increasing dimensionless velocities of withdrawal \( V = 0.0002, 0.002, 0.02, 0.2, 2 \). As expected, the viscoelastic peeling terminates only when contact radius is zero, and not at the JKR unstable radius. However, the minimum load is found for a contact radius which, for low velocities, is not too different from the unstable pull-off contact radius in JKR theory.
Fig. 3 - Load $P/P_0$ (a) and the contact radius $a/a_i$ (b) as a function of approach $\alpha/\alpha_i$. $P_0$ is JKR pull-off of the smooth sphere, and $\alpha_i$ the initial value of approach for unloading. The inner black wavy curve is the equilibrium Guduru solution, and the other 5 curves increasingly departing from it are obtained numerically for increasing dimensionless velocities of withdrawal (see the verse of the arrow) $V = 0.0002, 0.002, 0.02, 0.2, 2$. Here, $n = 0.33$, and other constants as indicated in Fig.1.

Fig. 4a,b,c give some detail of the solution at the lowest dimensionless velocity of withdrawal $V = 0.0002$. In particular, Fig. 4a shows clearly that the numerical solution follows closely the prediction of the Guduru elastic solution under displacement control, as expected, with almost sharp jumps
of the force at specific values of the approach. After the jump, the solution seems to return to the Guduru equilibrium solution. Obviously with the viscoelastic theory, the strict elastic solution should be obtained asymptotically at extremely low velocities, but the differential equation would then become very "stiff" corresponding to numerical difficulties following the jumps. The same behaviour is clarified in terms of the contact radius in Fig.4b which follows very closely the Guduru solution in some periods of time, then extends a little before jumping almost abruptly to the following branch of the equilibrium curve. In other words, the curve does not have a "rainflow" type of behaviour over the Guduru equilibrium solution, which would be the elastic real behaviour with jump-instabilities, but the contact radius "drops" over the Guduru curve only after some delays. This is further clarified in Fig.4c, where the velocity of the contact line $\frac{da}{d\alpha} = \frac{v_p}{v}$ is found to follow an oscillatory trend with "bursts" of very high (but finite) velocity where the peeling velocity is a much larger value than the imposed withdrawing velocity, after which the velocity drops to a low value which is where the contact area approaches the adiabatic Guduru curve since $G \simeq w_0$, and which increases progressively with the decreasing approach. Slowly, the solution departs from the Guduru elastic one, because of the cumulative effects of the acceleration periods. However, from Fig.3, and Fig.4d, for high velocities, we see that there are no real "jumps", and the solution curve is generally smoother, with the difference between the slow regime and the fast regime being smaller. Also notice in particular that while the velocity of peeling remains in every case equal to zero at the initial point, it remains closer to zero for a much extensive range of approach for high velocities, resulting in a curve departing away from the equilibrium Guduru curve immediately. This effect at high velocity produces curves that are generally closer to the viscoelastic curves for the smooth sphere, and therefore closer results for pull-off and work for pull-off. To see this from a quantitative point of view, since even the smooth sphere problem requires a numerical solution, we describe some results in the next paragraph.
Fig. 4 - Detail of the solution at the lowest dimensionless velocity of withdrawal (a,b,c) $V = 0.0002$, and (d) $V = 2$. Here, $n = 0.33$, and other constants as indicated in Fig. 1. In particular (a) Load-approach (b) contact radius vs approach (c) velocity of contact line $da/d\alpha = v_p/v$. (d) velocity of contact line $da/d\alpha = v_p/v$ but for the highest dimensionless speed $V = 2$.

3.2. Smooth sphere

We solve the problem for the smooth sphere under the same set of conditions (the equation for the smooth sphere are obviously obtained for $A = 0$ above), and obtained smooth results are shown in Fig. 5. Notice that as we discussed in the theory paragraph, in the initial point $da/d\alpha = 0$, and we find again (Fig. 5c) that the velocity remains practically zero for a longer time when $V$ is bigger. Here, contrary to the rough sphere, but also contrary to the smooth sphere in the case when load is constant, the velocity of the contact line increases monotonically from zero to infinite when pull-off occurs at zero contact area.
Fig. 5 - The load $P/P_0$ (a), the contact radius $a/a_i$ (b), and the velocity of contact line $da/d\alpha = v_p/v$ (c) as a function of approach $\alpha/\alpha_i$ for the smooth sphere. The inner black curve is the JKR classical solution, and the other 5 curves are obtained numerically for $V = 0.0002, 0.002, 0.02, 0.2, 2$ (follow the arrow). Here, $n = 0.33$, and other constants as indicated in Fig.1.

3.3. Some comparisons

Summarizing the pull-off results for $n = 0.33$, but adding some solutions also at different amplitudes of roughness $A$, we obtain the amplification factor for pull-off with respect to the JKR value as in Fig.6. Notice initially
that the smooth sphere results tend to a power-law scaling (linear in the log-log plot) as expected from the material law (1), after a transition from the elastic behaviour. As it is evident from the figure, starting off at low velocity with increasing amplitude of roughness, increases the "elastic" amplification according to the Guduru theory, but eventually the effect disappears at sufficiently large peeling speeds in the viscoelastic theory. In other words, there seems to be a "cross-over" between the two phenomena at the speed for which the two increases are the same. So in Fig.6, for example, for the very "rough" spheres of $A/\lambda = 0.045$ (blue line), we see that the Guduru enhancement is larger (7.5) that the viscoelastic one in this velocity range, and we don’t see a significant decrease of its effect, but eventually the smooth sphere result would be obtained for much larger speeds.

Obviously, with so many constants in the problem, it is not easy to give comprehensive results, but further tests with a larger value of $n = 0.6$ give results which confirm our conclusion about the crossover: namely pull-off force increases faster with the dimensionless speed factor $V$, but the Guduru effect disappears also faster.

![Fig.6](image_url)

Fig.6 - The pull-off amplification with respect to the JKR value, $P/P_0$ as a function of dimensionless speed of withdrawal $V$ for various amplitude of waviness increasing as indicated by arrow: $A/\lambda = 0$ for the smooth sphere (black), $A/\lambda = 0.005$ (green), $A/\lambda = 0.015$ (red), $A/\lambda = 0.045$ (blue). Here, all constants as indicated in Fig.1, except (a) $n = 0.33$; (b) $n = 0.6$.

4. Discussion

Considering the experiments on our geometry done by Guduru & Bull (2007), their gelatin material is indeed a viscoelastic material, as recognized
by Guduru & Bull (2007) who however did not characterized the material in particular and tried to minimize the loading rate effects by keeping in their tensile test machine a crosshead velocity at $v = 3mm/min = 50\mu m/s$ in all experiments. Indeed, even the smooth sphere case they use for measuring the baseline work of adhesion shows significant deviations between loading and the unloading curves which are not present in JKR theory. The unloading has specific features similar to what we found for the smooth sphere, namely when unloading begins, the contact radius does not begin to decrease immediately. Fitting JKR curves, Guduru & Bull (2007) extracted $w_0 = 0.008N/m$ during loading and $w'_0 = 0.22N/m$, a difference of a factor 27.5 but they used as baseline for their comparison with the wavy surfaces the unloading value. Despite experimental results capture generally the trend of the predictions, there is a ”systematic difference between the experimental observation and the theoretical prediction” as the authors say, of the order of a $-25\%$. We attribute this to the effect observed in the present paper, namely that since Guduru & Bull (2007) used their elastic theory with the work of adhesion already increased by viscoelastic effects, they overestimate the effect of load amplification.

Consider next the experiments by Barquins & Maugis (1981), where material was characterized: the viscoelastic toughness was measured with $n = 0.6$ in the range $v_p = 10^{-1} - 10^3\mu m/s$, i.e. over 4 decades of speed, which corresponds to an increase of 2 orders of magnitude in $\frac{w-w_0}{w_0}$. In ”tack” experiment of a 2.2$m$ glass ball on the polyurethane surface (i.e. pull-off experiments for which after a fixed contact time (5 min), at room temperature, a cross-head velocity is imposed in a tensile machine), an adherence force more than 30 times the quasistatic value at fixed displacement of the JKR theory, was found when $v = 1\mu m/s$. By increasing the cross-head speed by 4 orders of magnitude i.e. to $v = 1mm/s$, pull-off increased by a further factor of about 30. That is the viscoelastic effect can be very large, and therefore generally much larger than the Guduru effect. Also, Guduru effect only holds for a quite special waviness, and when the contact ”peels” quite uniformly around a circle, and requires the initial contact area to be compact, which poses some limits to the amplitude of roughness (see a more general numerical solution using Lennard-Jones force-separation law

\footnote{Measuring $v_p[\mu m/s]$, we have $\frac{w-w_0}{w_0} \simeq 10v_p^{0.6}$ from $w \simeq 3.5w_0$ to $w \simeq 632w_0$.}
Further, Li et al. (2019) numerical experiments for a rough sphere and elastic materials but with 2-dimensional wavy roughness (a generalization of the Guduru geometry which does not require the contact to peel in axisymmetric manner), the adhesion enhancement still persists although perhaps reduced (they found an increase of a factor 1.7 for a Johnson parameter $\alpha_{KLJ} = 0.37$ which corresponds to a Guduru 1D enhancement of $w = (1 + \frac{1}{\sqrt{\pi}}0.37)^2 = 6.37$)

and therefore the 2D sinusoidal waviness already decreases the Guduru effect significantly. Further numerical experiments in Li et al. (2019) with random roughness suggest that this enhancement was reduced to a maximum factor of 1.2, much smaller than expected from Guduru theory because of the random nature of roughness.

Returning finally to the experiments of Dalvi et al. (2019), the increase of apparent work of adhesion in the smoother specimen (Polished Ultra-NanoCrystalline Diamond (PUNCD)) is of a factor 2, at the retraction speed of 60nm/s. Given the discussed experimental findings of Barquins & Maugis (1981), which we can compare only qualitatively as the material may be quite different, if a factor 30 can be justified at $v = 1\mu m/s$, the factor 2 increase cannot be excluded not even at the 60nm/s speed. Regarding the roughness-induced enhancement, it was of a further factor less than 2, which could be either due to area increase as from the Persson-Tosatti (2001) theory, or from a reduced Guduru effect. But in view of the fact that the two enhancements are of the same order, this factor 2 may be too large, according to the present results and considering, to be attributed to Guduru’s effect, especially given the roughness is probably of random nature, and so assuming Li et al. (2019)’s results to be appropriate.

5. Conclusions

We have revisited the Guduru model for roughness-induced enhancement of adhesion of a sphere/flat contact, adding the effect of viscoelasticity which is expected in soft materials. The results have demonstrated that the roughness-induced amplification of pull-off in the Guduru model, which effectively can be modelled as an increased work of adhesion in the unloading
curve, is reduced progressively when velocity increases with respect to the baseline smooth viscoelastic sphere. This is also in qualitative agreement with the original experiments of Guduru and Bull (2007). A significant reduction has already occurred at a ”cross-over” velocity for which the two enhancement (the Guduru and the viscoelastic one) are of equal magnitude. We may be tempted therefore to speculate that viscoelasticity effectively damps the roughness-induced elastic instabilities, reduces roughness effects in unloading, while its effects are concentrated in the loading phase.

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7. References

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