Adhesion of viscoelastic media: an assessment of a recent JKR-like solution

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Abstract. Adhesion of soft compliant solids is irreversible and rate-dependent. As a result, two different paths are observed in loading-unloading adhesion experiments because of dissipation occurring in the unloading phase. An effective surface energy is usually introduced to take account of such dissipation.

Here, by exploiting a recent theoretical solution developed to study the detachment of a rigid sphere from a viscoelastic substrate (Violano et al., 2021), two different approaches are considered to calculate the surface energy. The first approach is based on the phenomenological equation derived by Gent & Schultz (A. N. Gent & J. Schultz, 1972), the latter exploits Persson & Brener theory for viscoelastic crack propagation (B. N. J. Persson & E. A. Brener, 2005).

In both cases, results are observed to be in good agreement with experimental data taken from the literature.

1. Introduction

"Adhesion of viscoelastic solids is irreversible and rate dependent. It presents a difficult problem in contact mechanics and is an ongoing field of research". This is an excerpt from a private letter - dated November 18, 2003 - written by K.L. Johnson. The recipient of the letter (V.L. Popov), recently presented it in the form of a note concerning the history of JKR theory [1].

Although eighteen years have passed since Johnson’s letter, adhesion of viscoelastic solids is still an open challenge for experts in contact mechanics and tribology. This is clearly demonstrated by recent efforts in experimental [2, 3], numerical [4, 6, 5] and theoretical works [7, 8].

In the contact between elastic spheres, Johnson, Kendall & Roberts (JKR) theory [9] predicts a pull-off force (i.e., the tensile load required for detaching the spheres) \( F_{PO} = \frac{1}{5} \pi \Delta \gamma R \), being \( R \) the equivalent radius of curvature of the spheres and \( \Delta \gamma \) the adiabatic value of the surface energy. However, experiments on soft materials (e.g., rubbers [10], elastomers [11], organic tissues [12]) have shown that the pull-off force increases by enhancing the unloading rate. As a result, the effective surface energy \( \Delta \gamma_{eff} \) observed in experiments is higher than its adiabatic value. Such behavior is related to rheological properties of soft matter.

In classical theoretical solutions [13, 14, 15], the debonding of a rigid indenter from a viscoelastic half-space is studied in the framework of Linear Elastic Fracture Mechanics (LEFM). The edge of contact line is seen as the tip of a propagating crack. Gent & Schultz (GS) [16] and Maugis & Barquins [13] (MB), first introduced phenomenological expressions to relate the effective surface energy with the crack tip velocity \( V_c \). In this class of theoretical models, two
common assumptions are done: i) debonding starts from a complete relaxed state of the material; ii) viscous dissipation is localized exclusively close to the crack tip.

Such assumptions are relaxed in recent numerical solutions [4, 6, 5]. Indeed, we developed a finite-element algorithm for studying the adhesion between a rigid sphere and a viscoelastic substrate [4]. When unloading starts from a relaxed state of the material, the pull-off force (and hence $\Delta \gamma_{\text{eff}}$) is found to monotonically increase with $V_c$ up to an asymptotic value reached at high unloading velocities. On the other hand, if unloading starts from an unrelaxed state of the material, the relation between pull-off force and $V_c$ takes the shape of a bell curve in a double-logarithmic plot.

Despite their potentiality and accuracy, numerical methods may require significant time and computational costs. For this reason, in a recent work [17], we proposed an extension of JKR theory to the contact between viscoelastic spheres. Such theoretical solution is here compared with the experimental outcomes of Ahn & Shull (AS) [11] by exploiting the phenomenological equation of Gent & Schultz to describe the dependence of $\Delta \gamma_{\text{eff}}$ on $V_c$. In addition, estimating $\Delta \gamma_{\text{eff}}$ from the viscoelastic crack propagation theory of Persson & Brener [19], a second comparison with the experimental data of Lorenz et al. [18] is proposed. Results show, in both cases, the model is quite in good agreement with reference data.

2. JKR-like viscoelastic model

The problem under investigation is sketched in Fig. 1.

![Figure 1](image_url)

**Figure 1.** The problem under investigation: a rigid spherical indenter pulled apart from a viscoelastic half-space.

The model developed in Ref. [17] assumes the half-space behaves as a linear elastic medium during the loading phase. In other words, loading is described by the classical JKR theory. Such condition can be reproduced in adhesion experiments by i) performing the loading phase at small driving velocity of the indenter [20], ii) waiting a sufficient long dwell time before unloading as complete relaxation of the viscoelastic substrate must be reached [2]. JKR theory [9] returns
the applied force $F$ and the penetration $\delta$ as functions of the contact radius $a$:

$$F = \frac{4}{3} \frac{E^* a^3}{R} - \sqrt{\frac{8\pi E^* a^3 \Delta \gamma}{a}}$$ (1)

$$\delta = \frac{a^2}{R} - \sqrt{\frac{2\pi a \Delta \gamma}{E^*}}$$ (2)

being $E^*$ the plain strain elastic modulus.

In order to extend JKR theory to viscoelastic contacts, we suggested (Ref. [17]) to replace the adiabatic surface energy $\Delta \gamma$ with its effective value $\Delta \gamma_{\text{eff}}$ in eqs. (1-2).

To estimate the effective surface energy we can exploit the phenomenological equation introduced by Gent & Schultz [16] and Maugis & Barquins [13]:

$$\Delta \gamma_{\text{eff}}(a) = \Delta \gamma [1 + \left(\frac{V_c(a)}{V^*}\right)^n]$$ (3)

where the viscoelastic parameters $V^*$ and $n$ can be measured experimentally [13, 2].

Alternatively, when the dependence of the viscoelastic modulus on the frequency of excitation is known, we can use Persson & Brener theory for viscoelastic cracks [19], according to which

$$\Delta \gamma_{\text{eff}}(a) = \Delta \gamma \left[1 - \frac{2E_0}{\pi} \int_0^{2\pi V_c(a)/s} d\omega \frac{F(\omega)}{\omega} \text{Im} \left(\frac{1}{E(\omega)}\right)\right]^{-1}$$ (4)

where $F(\omega) = |1 - (\omega s/(2\pi V_c))^2|^{1/2}$. In eq. (4), $s(V_c)$ is the crack tip radius and $E(\omega)$ is the viscoelastic modulus, being $E_0 = E(0)$ its value at vanishing excitation frequencies $\omega$. Moreover, PB showed that $s(V_c)/s_0 = \Delta \gamma_{\text{eff}}/\Delta \gamma$, where $s_0$ is the crack tip radius at vanishing contact line velocity and takes values of the order of 1 nm.

The contact line velocity is related to the contact radius $a$ by $V_c(a) = -da/dt$. If we assume that unloading occurs under fixed pulling velocity $V = -d\delta/dt$, then $V_c$ can be written as $V_c = V \cdot da/d\delta$.

To estimate $da/d\delta$, we initially assume constant $\Delta \gamma$ in eq. (2), so that the contact line velocity can be derived as

$$V_c(a) = V \left(\frac{2a}{R} - \sqrt{\frac{\pi \Delta \gamma}{2aE^*}}\right)^{-1}.$$ (5)

Substituting the above relation, in eq. (3) or (4) one can calculate the effective surface energy $\Delta \gamma_{\text{eff}}$, which in turn can be used in JKR eqs. (1-2) to obtain force and penetration in the viscoelastic case.

Experiments on soft materials [2, 21, 22] show the existence of a *stick zone* during the initial phase of debonding. Therefore, according to Ref. [17], in the initial stick phase, we assume the spherical indenter behaves as a circular flat punch of radius equal to the value $a_{\text{max}}$ reached at the end of loading. As a result, we have

$$\delta = \delta_{\text{max}} - \frac{F_{\text{max}} - F}{2E^* a_{\text{max}}}.$$ (6)

being, $F_{\text{max}}$ and $\delta_{\text{max}}$ the force and the penetration at the beginning of unloading, respectively.

Eq. (6) holds for $\delta_c \leq \delta \leq \delta_{\text{max}}$, being $\delta_c$ the critical penetration over which $a$ starts to decrease. On the contrary, for $\delta < \delta_c$, contact is described by eqs. (1-2) with $\Delta \gamma$ replaced by $\Delta \gamma_{\text{eff}}$. 
3. Results

A first comparison of the predictions of the described model is proposed with the experimental data collected by Ahn & Shull (AS) [11], which carried out JKR adhesion tests between a model acrylic elastomer and a glassy polymeric substrate. Specifically, hemispherical lenses of lightly cross-linked PolyN-ButylAcrylate (PNBA) with radius of curvature $R = 1\, \text{mm}$ were subjected to loading/unloading cycles on flat PolyMethylMethAcrylate (PMMA) substrates.

In AS experiments, loading phase is performed by increasing the load step by step and waiting a fixed time at the end of each step. Moreover, before unloading, the maximum load $F_{\text{max}}$ is maintained constant for a dwell time of 10 min. In such case, quasi-static conditions are ensured (see Ref. [23] for explanation) and JKR solution can be used to estimate the elastic modulus and the adiabatic surface energy (for AS experiments we find $E^* = 0.17\, \text{MPa}$ and $\Delta\gamma = 22\, \text{mJ/m}^2$). During unloading, the contact line velocity $V_c = -da/dt$ is obtained by capturing pictures of the contact area at fixed frame rates.

Figure 2 shows the trend of $V_c$ with the contact radius $a$, for different pulling velocities $V = -d\delta/dt$. Markers refer to experimental data from Ref. [11], while solid lines are the approximate solutions returned by eq. (5). Unfortunately, the values of $V$ are not reported in Ref. [11], so we have used $V$ as a fitting parameter in eq. (5). Moreover, we have assumed that unloading occurs at constant $V$.

![Figure 2.](image)

Following Maugis & Barquins (MB) [13], AS calculated the energy release rate $G$ with

$$G = \frac{(F_H - F)^2}{6\pi RF_H},$$

(7)

where $F_H = 4E^*a^3/(3R)$ is the Hertzian load and $F$ is the applied load (known from experiments).
Figure 3. The relative increase in the effective surface energy \((\Delta\gamma_{\text{eff}} - \Delta\gamma) / \Delta\gamma\) in terms of the contact line velocity \(V_c\). Markers refer to experimental data from Ref. [11], while solid line is the fit obtained by means of eq. (3) with \(V^* = 2.24 \times 10^{-7} \text{ m/s}\) and \(n = 0.83\).

In the framework of LEFM [13], at equilibrium \(G = \Delta\gamma_{\text{eff}}\). Therefore, experimental data can be used to fit the values of the parameters \(V^*\) and \(n\) in eq. (3), as shown in Fig. 3, where the relative increase in the effective surface energy \((\Delta\gamma_{\text{eff}} - \Delta\gamma) / \Delta\gamma\) is given in terms of the contact line velocity \(V_c\). Again, markers refer to experimental data, while solid line is the fit obtained by eq. (3), which returns \(V^* = 2.24 \times 10^{-7} \text{ m/s}\) and \(n = 0.83\).

Figure 4. The contact radius \(a\) in terms of the applied load \(F\). Markers refer to experimental data from Ref. [11], while lines denote theoretical predictions. Results are shown for different pulling velocities \((V = 0.28, 0.8, 2.3, 7.2 \mu\text{m/s})\).
Figure 5. The pull-off force $F_{PO}$ as a function of the pulling velocity $V$. Red squares refer to experimental data from Ref. [11], while green triangles are theoretical predictions.

Figure 4 shows the curves relating the contact radius $a$ with the contact force $F$ for increasing pulling velocities $V$. Black empty circles are the experimental data collected in the loading phase, for which viscoelastic effects are negligible. JKR elastic theory (black dashed line) is clearly working in this case. Colored markers refer instead to the experimental data collected during unloading at different $V$. Adhesion hysteresis occurs as loading and unloading paths are different and the stick zone is observed to increase with the pulling velocity. Results of the theoretical model (colored solid lines) match quite well the experiments, even if at high pulling rates the stick time is slightly overestimated.

Figure 6. The real and imaginary part of the viscoelastic modulus $E(\omega)$ of the PDMS used in the experiments of Ref. [18].
Figure 7. The effective surface energy $\Delta \gamma_{\text{eff}} / \Delta \gamma$ in terms of the contact line velocity $V_c$ for the PDMS sample used in Ref. [18]. Equation (4) has been used for the calculation of $\Delta \gamma_{\text{eff}}$.

The pull-off force, i.e., the maximum tensile (negative) load measured during unloading, can be calculated by solving the equation $dF/da = 0$. In figure 5 we compare the measured pull-off force with the theoretical predictions, for increasing pulling velocity $V$. In such case, a very good agreement is observed also at relatively high rates.

Figure 8. The contact radius $a$ in terms of the applied load $F$. Markers refer to experimental data from Ref. [18], while lines denote theoretical predictions. Results are shown for different radius of curvature $R = 1.6, 2.4$ mm and pulling velocity $V = 0.1 \, \mu\text{m/s}$.

The theoretical predictions of the model are also compared with the experimental findings given in Lorenz et al. [18], where an optical microtribometer is used to perform loading-unloading experiments between polished silicon nitride spheres and a flat PolyDiMethylSiloxane (PDMS) elastomer. Lorenz et al. [18] also measured the viscoelastic modulus $E(\omega)$ of PDMS in a wide
range of frequencies of excitation (see Fig. 6). So, the effective surface energy can be calculated by eq. (4), and its dependence on contact line velocity $V_C$ is shown in Fig. 7.

Figure 8 shows the loading-unloading curves relating $a$ and $F$ for two values of the radius of the spheres used in the experiments (namely $R = 1.6$ mm and $R = 2.4$ mm). Loading data (empty markers) are fitted using JKR theory (dashed lines), which returns $E^*=2.55$ MPa and $\Delta \gamma = 46$ mJ/m$^2$. Unloading (filled markers) is assumed to be performed at $V = 0.1 \mu$m/s according to Ref. [24]. Once again, the theoretical predictions (solid lines) closely match the experimental data. Also, notice an increase in the sphere radius results in an increase of the dissipated energy (wider area between the loading and unloading curves) as the region involved by viscous effects becomes larger.

4. Conclusions
In this work, JKR-like experiments performed between soft bodies are interpreted in the light of a very simple theoretical model, where viscous effects are considered by replacing in JKR equations the adiabatic surface energy $\Delta \gamma$ with the effective value $\Delta \gamma_{\text{eff}}$ which takes account of dissipation.

First, the predictions of the model are compared with the experimental findings of Ref. [11]. In such case, the effective surface energy is calculated by the phenomenological equation proposed by Gent & Schultz (GS) [16] and Maugis & Barquins [13].

A second set of experimental data, taken from Lorenz et al. [18], is then used to test the model when $\Delta \gamma_{\text{eff}}$ is estimated according to Persson & Brener theory for viscoelastic cracks [19].

In general, notwithstanding its simplicity, the proposed model correctly captures the rate-dependent behaviour occurring in the unloading phase. However, we stress the model is strictly valid when viscoelastic dissipation occurs close to the contact line edge, i.e., under the assumption that the bulk material behaves elastically.

[1] Popov V L 2021 A Note by KL Johnson on the History of the JKR Theory. Tribology Letters 69(4) 1-3.
[2] Violano G, Chateauminois A and Afferrante L 2021 Rate-dependent adhesion of viscoelastic contacts, Part I: Contact area and contact line velocity within model randomly rough surfaces. Mechanics of Materials 160 103926.
[3] Das D and Chasiotis I 2021 Rate dependent adhesion of nanoscale polymer contacts. Journal of the Mechanics and Physics of Solids 156 104597.
[4] Afferrante L and Violano G 2021 On the effective surface energy in viscoelastic Hertzian contacts. arXiv preprint arXiv:2107.03796.
[5] Van Dokkum J S, Pérez-Rafols F, Dorogin L and Nicola L 2021 On the retraction of an adhesive cylindrical indenter from a viscoelastic substrate. Tribology International, 164 107234.
[6] Müser M H and Persson B N 2021 Crack and pull-off dynamics of adhesive, viscoelastic solids. arXiv preprint arXiv:2108.02031.
[7] Ciavarella M 2021 An upper bound for viscoelastic pull-off of a sphere with a Maugis-Dugdale model. The Journal of Adhesion, 1-14.
[8] Persson B N J 2021 A simple model for viscoelastic crack propagation. The European Physical Journal E 44(1) 1-10.
[9] Johnson K L, Kendall K and Roberts A 1971 Surface energy and the contact of elastic solids. Proceedings of the royal society of London. A. mathematical and physical sciences 324(1558) 301-313.
[10] Roberts A D 1979 Looking at rubber adhesion. Rubber Chemistry and Technology 52(1) 23-42.
[11] Ahn D. and Shull K R 1996 JKR studies of acrylic elastomer adhesion to glassy polymer substrates. Macromolecules 29(12) 4381-4390.
[12] Han G, Eriten M and Henak C R 2020 Rate-dependent adhesion of cartilage and its relation to relaxation mechanisms. Journal of the mechanical behavior of biomedical materials 102 103493.
[13] Maugis D and Barquins M 1980 Fracture mechanics and adherence of viscoelastic solids. In Adhesion and adsorption of polymers pp. 203-277. Springer, Boston, MA.
[14] Greenwood J A and Johnson K L 1981 The mechanics of adhesion of viscoelastic solids. Philosophical Magazine A 43(3) 697-711.
[15] Muller V M 1999 On the theory of pull-off of a viscoelastic sphere from a flat surface. Journal of Adhesion Science and Technology 13(9) 999-1016.
[16] Gent A N and Schultz J 1972 Effect of wetting liquids on the strength of adhesion of viscoelastic material. The Journal of Adhesion 3(4) 281-294.
[17] Violano G, Chateauminois A and Afferrante L 2021 A JKR-like solution for viscoelastic adhesive contacts. Frontiers in Mechanical Engineering 7 25.
[18] Lorenz B, Krick B A, Mulakaluri N, Smolyakova M, Dieluweit S, Sawyer W G and Persson B N J 2013 Adhesion: role of bulk viscoelasticity and surface roughness. Journal of Physics: Condensed Matter 25(22) 225004.
[19] Persson B N J and Brener E A 2005 Crack propagation in viscoelastic solids. Physical Review E 71(3) 036123.
[20] Dalvi S, Gujrati A, Khanal S R, Pastewka L, Dhinojwala A and Jacobs T D 2019 Linking energy loss in soft adhesion to surface roughness. Proceedings of the National Academy of Sciences 116(51) 25484-25490.
[21] Baek D, Hemthavy P, Saito S and Takahashi K 2017 Evaluation of energy dissipation involving adhesion hysteresis in spherical contact between a glass lens and a PDMS block. Applied Adhesion Science 5(1) 1-11.
[22] Deruelle M, Hervet H, Jandeau G, and Léger L 1998 Some remarks on JKR experiments. Journal of adhesion science and technology 12(2) 225-247.
[23] Acito V, Ciavarella M, Prevost A M, and Chateauminois A 2019. Adhesive contact of model randomly rough rubber surfaces. Tribology Letters 67(2) 54.
[24] Krick B A 2021 Private communication.