



On the Long and Short-Range Adhesive Interactions in Viscoelastic Contacts

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Abstract

Recently, tribologists have shown increasing interest in rate-dependent phenomena occurring in viscoelastic fractures. However, in some cases, conflicting results are obtained despite the use of similar theoretical models. For this reason, we try to shed light on the effects that long and short-range adhesion has on the pull-off force in the contact of viscoelastic media by exploiting a recently developed numerical model. We find that, in the limit of long-range adhesion, the unloading velocity has little effect on the pull-off force, which is close to the value predicted by Bradley for rigid bodies. In such case, the detachment process is characterized by a uniform bond-breaking of the contact area, and viscous dissipation involves the bulk material. For medium(short)-range adhesion, the pull-off force is instead a monotonic increasing function of the pulling velocity and, at high speeds, reaches a plateau that is a function of the adiabatic surface energy. In this case, the detachment process is similar to the opening of a circular crack, and viscous dissipation is localized at the contact edge.

Keywords Rate-dependent adhesion · Long and short-range adhesion · Tabor parameter · Finiteelement method · Pull-off force

1 Introduction

In the 1970s, two pioneering contact mechanics theories were formulated to study the adhesion between elastic spheres. First, Johnson, Kendall and Roberts (JKR) [1] exploited a thermodynamic approach to include adhesion in the Hertz theory. JKR solution assumes infinitely short-range adhesive interactions inside the contact area, that cause a deformation of the Hertzian contact profile. Later, Derjaguin, Muller and Toporov (DMT) [2] proposed a different solution where weak long-range adhesive interactions are localized outside the contact area, without deforming the Hertzian gap.

Both JKR and DMT theories predict a pull-off force, i.e., the maximum tensile force reached during the detachment that is independent of the elastic properties of the half-space and equal to $1.5\pi\Delta\gamma R$ and $2\pi\Delta\gamma R$, respectively, being R the radius of the sphere and $\Delta\gamma$ the adiabatic surface energy.

Tabor [3] first clarified that JKR theory is accurate in the limit of high values of a dimensionless parameter μ (known as Tabor parameter), namely for soft materials with high surface energy and radii of curvature. On the contrary, DMT theory is accurate in the limit of $\mu \ll 1$. Notice DMT pull-off force is the same calculated by Bradley [4] in the limit of rigid bodies.

Maugis [5], exploiting the Dugdale cohesive law, formulated a theory, known as Maugis-Dugdale (MD) theory, which allows to capture the JKR-DMT transition in terms of μ . Therefore, he showed that, moving from JKR to DMT limit, the pull-off force ranges in between $1.5\pi\Delta\gamma R$ and $2\pi\Delta\gamma R$, as also confirmed by a successive numerical study [6].

In modern adhesive systems, soft materials are widely used to enhance adhesive features. Such materials exhibit rate-dependent adhesion as a consequence of their intrinsic viscoelasticity, while JKR, DMT, and MD theories cannot capture rate effects and viscous dissipation. In fact, experiments have shown that the viscoelastic pull-off force may be order of magnitudes larger than the value predicted by *elastic* adhesion theories (see, for example, Ref. [7]).

Recently, Das and Chasiotis (DC) [8] proposed to extend MD model to the case of a rigid sphere indenting a

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viscoelastic half-space. In their works, unloading starts from a relaxed state of the viscoelastic material and is performed at fixed rate of the applied force.

In the limit of short-range adhesive interactions, DC model predicts the pull-off force to be a monotonic increasing function of the unloading rate. If $F_{PO,0}$ denotes the elastic pull-off force, the maximum pull-off force that can be reached at high unloading rate is $F_{PO,max} = (F_{PO,0}E_{\infty})/E_0$, being E_{∞} and E_0 the instantaneous and relaxed moduli of the viscoelastic material, respectively. However, such value cannot be reached for any value of the Tabor parameter (see, for example, Refs. [9–11]).

Moreover, when long-range adhesive interactions are predominant, namely when $\mu \rightarrow 0$, DC model continues to predict $F_{PO,max} = (F_{PO,0}E_{\infty})/E_0$ at high unloading rate, while in Ref. [11] it is observed that $F_{PO,max} \rightarrow F_{PO,0}$ independently of the value of the unloading rate.

In this work, we try to clarify such qualitative and quantitative discordance by performing contact simulations with the fully deterministic model developed in Ref. [12], where adhesive interactions are modeled with Lennard-Jones-based potential and viscoelasticity with the standard linear solid.

2 Statement of the Problem

Consider a rigid sphere, with radius of curvature R , pulled off from a viscoelastic substrate. Detachment starts from static equilibrium conditions as the sphere initially approaches the substrate at low speed up to a maximum penetration δ_{max} . Retraction is then performed at a constant speed $V = -d\delta/dt$.

The contact problem is solved using the finite element (FE) model presented in Ref. [12], at which the reader is referred for further details. We here recall that: (i) interface interactions are modelled by a traction-gap law based on Lennard-Jones potential, according to the *proximity approximation* (i.e., the force acting between two bodies is expressed in terms of the force between two semi-infinite planes); (ii) the viscoelastic behaviour of the substrate is modelled with the standard linear solid according to the Maxwell representation.

We are interested in studying how the pull-off force is affected by the retraction speed V , in the cases of short, medium, and long-range adhesive interactions. In the elastic case, the Tabor parameter is defined as

$$\mu = \frac{R^{1/3}}{\epsilon} \left[\frac{\Delta\gamma(1 - \nu^2)}{E_0} \right]^{2/3}, \tag{1}$$

where E_0 is Young’s modulus of the substrate, ϵ is the range of action of attractive forces, and ν is Poisson’s ratio.

For a viscoelastic material, Young’s modulus $E(\omega)$ is a function of the frequency of excitation and hence varies during the detachment process. Viscoelastic modulus $E(\omega) \rightarrow E_0$ only when detachment occurs in quasi-static conditions, namely when $V \rightarrow 0$. As a result, while in the elastic case we are sure that μ uniquely describes the trend of the pull-off force, in the viscoelastic case this it is not necessarily true. However, we can reasonably assume that long-range adhesion is expected for low values of the adiabatic surface energy $\Delta\gamma$, resulting in $\mu \ll 1$, while medium and long-range adhesion occurs at higher $\Delta\gamma$. In the following, we shall discuss our results in terms of $\Delta\gamma$ rather than μ .

3 Results and Discussion

All plots are given for $R/\epsilon = 500$, $E_{\infty}/E_0 = 10$ and $\nu \approx 0.5$, and in terms of the following dimensionless quantities: $\hat{F}_{PO} = F_{PO}/(\pi\Delta\gamma R)$; $\Delta\hat{\gamma} = \Delta\gamma/(E_0R)$; $\hat{V} = V\tau/\epsilon$, being τ the relaxation time of the viscoelastic material. Furthermore, the unloading phase started after reaching the penetration $\hat{\delta}_{max} = \delta/\epsilon = 17.7$. Yu and Polycarpou [13] showed that, for the Lennard-Jones potential law, the ‘exact’ value of ϵ is affected by material properties, such as the interfacial surface energy. Here, we fixed $\epsilon = 0.3$ nm, assuming it is independent of $\Delta\gamma$. This assumption is usually adopted in adhesive contact simulations [14]. However, even considering a dependence of ϵ with $\Delta\gamma$, the qualitative trend of the pull-off force with the surface energy is not expected to change, while some quantitative differences may occur [13].

Figure 1 shows the normalized pull-off force \hat{F}_{PO} in terms of the pulling velocity \hat{V} , which is constant during the retraction of the sphere. Results are obtained for different values of the adiabatic surface energy $\Delta\hat{\gamma}$ and for \hat{V} ranging from

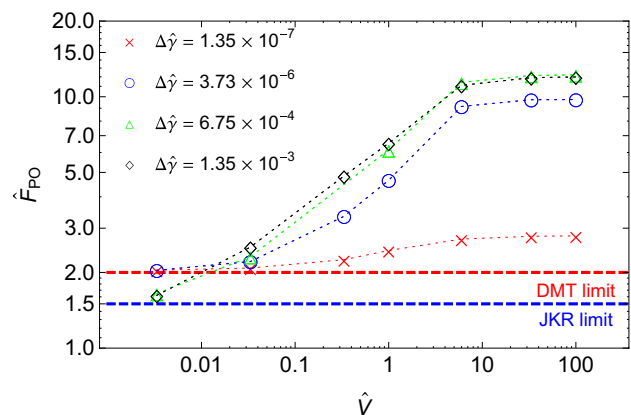


Fig. 1 The normalized pull-off force \hat{F}_{PO} in terms of the dimensionless pulling velocity \hat{V} . Finite element calculations (markers) are shown for $\Delta\hat{\gamma} = 1.35 \times 10^{-7}$, 3.73×10^{-6} , 6.75×10^{-4} , 1.35×10^{-3} . DMT and JKR elastic limits are also shown as a reference

0.0033 to 100. First, let us consider what happens when $V \sim 0$; for the lower values of $\Delta\hat{\gamma}$, the DMT limit is returned, being $\hat{F}_{PO} \sim 2$. As a result, adhesion is basically described by long-range interactions. On the contrary, for the higher values of $\Delta\hat{\gamma}$, $\hat{F}_{PO} \sim 1.5$ and the JKR limit is recovered being dominant the short-range adhesion. Notice, the JKR limit is not closely reached as, in our calculations, finite values of the Tabor parameter are considered [10]. In all cases, \hat{F}_{PO} is a monotonic increasing function of \hat{V} and reaches a maximum asymptotic value $\hat{F}_{PO,max}$ at high pulling velocities. However, notice $\hat{F}_{PO,max}$ is strongly affected by the value of $\Delta\hat{\gamma}$.

In this regard, fracture mechanics and cohesive zone models (see, for example, Refs. [15, 16]) suggest that $F_{PO,max}/F_{PO,0} = E_\infty/E_0$, being $F_{PO,0}$ the pull-off force in the elastic limit. Our calculations, instead, returns $F_{PO,max}/F_{PO,0} \sim 0.8E_\infty/E_0$ for $\Delta\hat{\gamma} = 1.35 \times 10^{-3}$ and $F_{PO,max}/F_{PO,0} \sim 0.14E_\infty/E_0$ for $\Delta\hat{\gamma} = 1.35 \times 10^{-7}$. These results agree with the theoretical findings given in Ref. [11], where it is suggested that there is an upper bound of the pull-off force that depends on the Tabor parameter. However, this is not the only reason for which $F_{PO,max}/F_{PO,0}$ is less than E_∞/E_0 ; indeed, in a recent study [9], we have shown that geometric and finite-size effects influence $F_{PO,max}$, which decreases when retraction starts from smaller initial contact radii a_{max} .

To highlight the effect of the adiabatic surface energy on the maximum pull-off force, Fig. 2 shows $\hat{F}_{PO,max}$ in terms of $\Delta\hat{\gamma}$ and for $\hat{V} = 100$. Results disagree with DC findings [8], where $\hat{F}_{PO,max}$ is predicted to increase when we move from short-range to long-range adhesive interactions for all rates of unloading (see Fig. 6C in Ref. [8]).

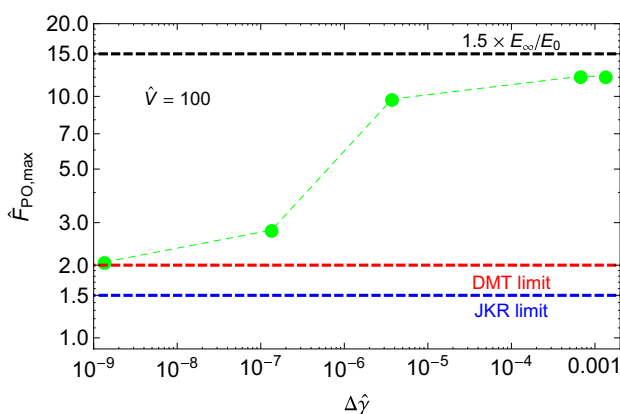


Fig. 2 The maximum pull-off force $\hat{F}_{PO,max}$ in terms of the dimensionless adiabatic surface energy $\Delta\hat{\gamma}$. $\hat{F}_{PO,max}$ is calculated at high pulling speed ($\hat{V} = 100$). Green markers refer to finite element calculations, red and blue dashed lines are DMT and JKR elastic limits, respectively, and black dashed line is the theoretical limit predicted at high speeds by fracture mechanics models

Therefore, in DC calculations, $\hat{F}_{PO,max}$ is surprisingly found to be a decreasing function of the Tabor parameter.

However, we have to observe that our outcomes agree with results given in Ref. [11], where a similar model to DC one, based on Maugis–Dugdale approach, is adopted. Our results are also in agreement with recent numerical findings of Muser and Persson (MP) [10], who performed calculations on the detachment of a rigid cylinder from a viscoelastic substrate. They found a decrease in $\hat{F}_{PO,max}$ when reducing the Tabor parameter μ . However, they changed the size of the radius R of the indenter maintaining the same value of $\Delta\hat{\gamma}$. In such conditions, size effects due to a_{max} may arise, leading to additional variations in the pull-off force [9]; moreover, at small scales, a transition of the detachment mode from crack propagation to uniform bond-breaking can also occur [9, 10, 17].

In addition, Van Dokkum et al. [18] observed, in a similar problem, the normalized effective surface energy $\Delta\gamma_{eff}/\Delta\gamma$ (or equivalently \hat{F}_{PO}) decreases with the Tabor parameter. Furthermore, Jiang et al. [19], performing FE calculations on the pull-off force of a spherical indenter from a soft viscoelastic stamp, observed a reduction of \hat{F}_{PO} when decreasing $\Delta\hat{\gamma}$. Similarly to our approach, they changed the value of $\Delta\hat{\gamma}$ maintaining the same maximum penetration δ_{max} .

Our results find confirmation also in the experimental measurements of Ahn and Shull (AS) [20], who performed axisymmetric adhesion tests between a hemispherical elastomeric cap and a variety of flat substrates with different values of interface energy. Moving from the JKR formalism, they used a standard fracture mechanics approach to estimate the effective surface energy $\Delta\gamma_{eff}$ according to the phenomenological equation $\Delta\gamma_{eff} = \Delta\gamma[1 + f(V_c)]$ [21], where the quantity $f(V_c)$ takes into account the increase in surface energy due to viscous dissipation and depends only on the contact line velocity $V_c = -da/dt$ (provided the process is assumed occurring at constant temperature).

Figure 3A, B show the increase in the dissipated energy density ΔU_D at pull-off, for $\hat{V} = 1$ and $\Delta\hat{\gamma}$ equal to 1.35×10^{-3} and 1.35×10^{-7} , respectively. For high values of the adiabatic surface energy (Fig. 3A), viscous dissipation is localized at the edge of contact, suggesting that the detachment process is similar to the mechanism of opening of a circular crack [12]. For low $\Delta\hat{\gamma}$ (Fig. 3B), a much lower viscous dissipation is observed. It mainly involves the bulk material, suggesting the occurrence of a different debonding mechanism, as clarified in Fig. 4A, B, where the normal displacement (net of the rigid one) is shown at three different times close to the pull-off instant (namely the instant at which the tensile force is maximum).

Results are given for the two cases corresponding to Fig. 3A, B. For $\Delta\hat{\gamma} = 1.35 \times 10^{-3}$, the debonding

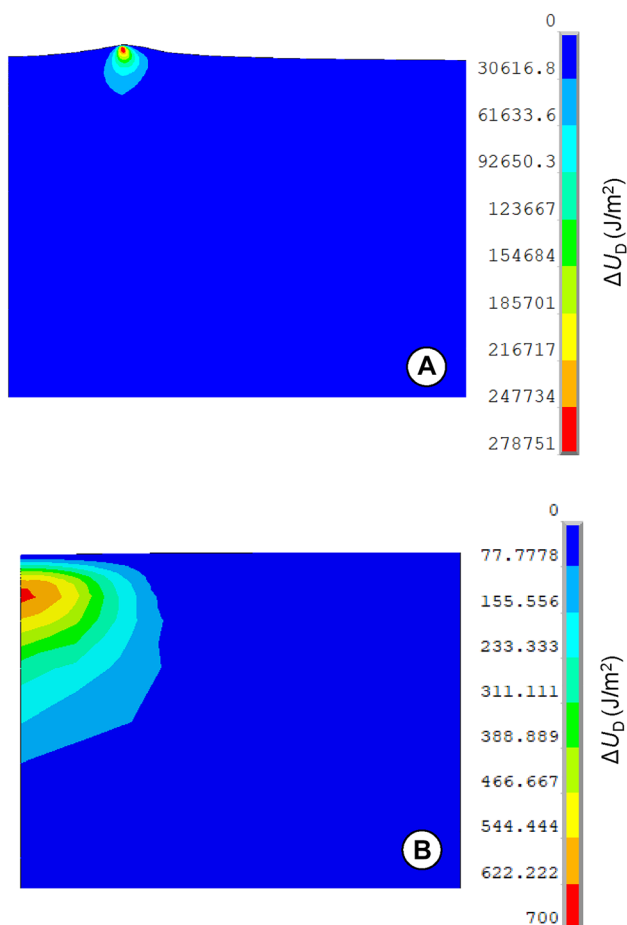


Fig. 3 The increase in the dissipated energy density at pull-off. Results are given for a pulling speed $\hat{V} = 1$, with $\Delta\hat{\gamma} = 1.35 \times 10^{-3}$ (A) and $\Delta\hat{\gamma} = 1.35 \times 10^{-7}$ (B)

mechanism is clearly related to crack propagation; for $\Delta\hat{\gamma} = 1.35 \times 10^{-7}$, the displacement field instead moves homogeneously leading to a quasi-uniform bond-breaking.

4 Conclusions

In this paper, we investigate the effect of the adhesive interactions on the pull-off force of viscoelastic bodies when the detachment process starts from a relaxed equilibrium state of the material.

In the limit of long-range adhesion ($\Delta\gamma \rightarrow 0$), quasi-uniform bond-breaking describes the mechanism of detachment, and viscous dissipation occurs in the bulk material. As a result, the pull-off force is not affected by the pulling speed and tends to the rigid limit of Bradley [4].

When medium(short)-range interactions occur, the detachment process is similar to that of a propagating

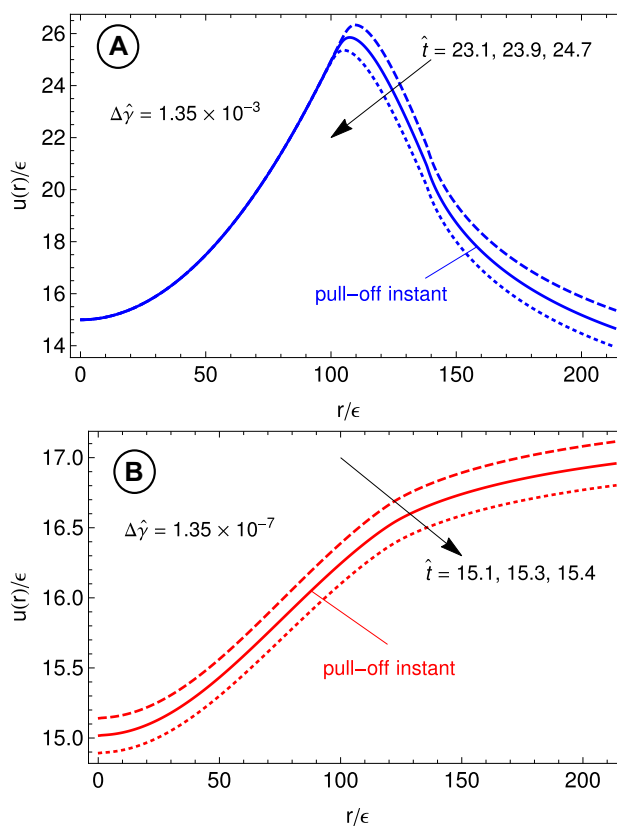


Fig. 4 Displacement fields in the vicinity of the pull-off instant, for a pulling speed $\hat{V} = 1$, with $\Delta\hat{\gamma} = 1.35 \times 10^{-3}$ (A) and $\Delta\hat{\gamma} = 1.35 \times 10^{-7}$ (B). The dimensionless time $\hat{t} = t/\tau$ increases in the direction of the arrow. The displacements are given net of the rigid one

circular crack. In this case, the pull-off force is a monotonic increasing function of the pulling speed. At high speeds, the pull-off load reaches a plateau, which depends on $\Delta\gamma$.

Therefore, we can state that the pull-off force is limited by the strength of adhesion ($\Delta\gamma$), rate of unloading and finite-size effects (a_{\max}). This is the reason why the ratio between the actual pull-off force and its elastic value does not approach the theoretical limit E_∞/E_0 at high pulling speeds.

Such outcomes also have important implications in problems of viscoelastic fracture mechanics.

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Declarations

Conflict of interest The authors declare that there are no conflicts of interests within the outlined work.

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