



Bulk and fracture process zone contribution to the rate-dependent adhesion amplification in viscoelastic broad-band materials

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ARTICLE INFO

Dataset link: <https://doi.org/10.5281/zenodo.13358696>

Keywords:

Adhesion
Viscoelasticity
Sphere contact
Enhancement
Pull-off
Modified power law
Surface energy

ABSTRACT

The contact between a rigid Hertzian indenter and an adhesive broad-band viscoelastic substrate is considered. The material behavior is described by a modified power law model, which is characterized by only four parameters, the glassy and rubbery elastic moduli, a characteristic exponent n and a timescale τ_0 . The maximum adherence force that can be reached while unloading the rigid indenter from a relaxed viscoelastic half-space is studied by means of a numerical implementation based on the boundary element method, as a function of the unloading velocity, preload and by varying the broadness of the viscoelastic material spectrum. Through a comprehensive numerical analysis we have determined the minimum contact radius that is needed to achieve the maximum amplification of the pull-off force at a specified unloading rate and for different material exponents n . The numerical results are then compared with the prediction of Persson and Brener viscoelastic crack propagation theory, providing excellent agreement. However, comparison against experimental tests for a glass lens indenting a PDMS substrate shows data can be fitted with the linear theory only up to an unloading rate of about 100 $\mu\text{m/s}$ showing the fracture process zone rate-dependent contribution to the energy enhancement is of the same order of the bulk dissipation contribution. Hence, the limitations of the current numerical and theoretical models for viscoelastic adhesion are discussed in light of the most recent literature results.

1. Introduction

Understanding the adhesive behavior of soft materials such as polymers and elastomers would be of interest in many engineering applications, ranging from friction (Lorenz et al., 2015; Peng et al., 2021; Nazari et al., 2024; Mandriota et al., 2024), gripping technologies (Shintake et al., 2018), switchable adhesion (Kamperman et al., 2010; Papangelo and Ciavarella, 2017; Linghu et al., 2023, 2024), bio-mechanics (Felicetti et al., 2022; Forsbach et al., 2023), and soft robotics (Mazzolai et al., 2019; Agnelli et al., 2021). Since the fundamental contribution of Johnson, Kendall, and Roberts, the JKR contact model (Johnson et al., 1971), it is known that adhesion of compliant elastic materials can be understood as a Griffith energy balance between the elastic strain energy released as the crack advances and the energy dissipated by the formation of new surfaces, as it is classically known in Linear Elastic Fracture Mechanics (Maugis, 1992). In this case, for quasi-static conditions, a thermodynamic work of adhesion $\Delta\gamma_0$, sometimes referred to as “surface energy”, can be defined that is independent of the rate at which the remote load is applied.

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<https://doi.org/10.1016/j.jmps.2024.105844>

Received 30 June 2024; Received in revised form 23 August 2024; Accepted 1 September 2024

Available online 2 September 2024

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If the crack is advancing in a viscoelastic media, care should be taken in accounting for the dissipative effects introduced by viscoelasticity. Currently, few theories attempt a description of this problem. The cohesive zone models (Schapery, 1975a,b; Greenwood and Johnson, 1981; Greenwood, 2004; Schapery, 2022b) attempt an accurate description of the adhesive interactions taking place at the crack mouth, more precisely within a “process zone” of length l_0 where the material bonds are actually disrupted and which is a function of the crack speed, resulting in the dependence on the speed of the effective surface energy $\Delta\gamma_{eff}$. A different model developed by Persson and Brener (2005) (PB theory in the following) focuses on a steady-state moving crack. It establishes an energy balance between the power supplied by the applied load and the power dissipated by viscoelastic losses within the bulk material and the creation of new surfaces. By introducing a critical stress threshold for the rupture of material bonding, σ_c , it also introduces a typical lengthscale l_0 to calculate the dissipation in the bulk viscoelastic material (Persson and Brener, 2005; Persson, 2017, 2021).

Both cohesive and dissipation models really require a reference stress, or a reference length scale, which ultimately is used as a free parameter to fit the data points, and the linear theories result sometimes in nonphysical size for the fracture process zone at low speeds (Hui et al., 2022), as we shall discuss with reference to our results in the Discussion paragraph. Schapery developed also more elaborate theories (Schapery, 1984) to include nonlinear stress–strain behavior using J integral and also far-field viscoelasticity, which require obviously even more material characterization.

For a semi-infinite system, the cohesive-model approach and PB theory lead to a monotonic increase of the effective (or “apparent”) surface energy $\Delta\gamma_{eff}$ with the velocity v up to the theoretical “high-frequency” limit of $\Delta\gamma_{eff}/\Delta\gamma_0 = E_\infty/E_0$ being E_∞ and E_0 respectively the glassy and the rubbery elastic moduli of the viscoelastic material (Ciavarella et al., 2021). For the unloading of a flat punch from a viscoelastic substrate, both approaches have been recently revisited to include finite-size effects, which have been showed to still give a monotonic increase of the effective surface energy, but with a maximum amplification that is limited by the system dimension providing $\max(\Delta\gamma_{eff}/\Delta\gamma_0) < E_\infty/E_0$ (Maghami et al., 2024). Even for systems that can be considered semi-infinite, the contact problem presents several challenges as macroscopic adhesion is generally influenced by the indenter geometry (Papangelo and Ciavarella, 2023; Maghami et al., 2024) and the contact history (Greenwood and Johnson, 1981; Violano et al., 2021; Violano and Afferrante, 2022; Afferrante and Violano, 2022; VanDonselaar et al., 2023).

To precisely assess adhesion in soft polymers (silicone, rubber), the properties of the viscoelastic material need to be characterized. Several numerical and experimental works have tried to accurately determine the viscoelastic material response in the time domain Wayne Chen et al. (2011), Lin et al. (2022), Dusane et al. (2023), Qi et al. (2024), in the frequency domain Huang et al. (2004), Efremov et al. (2017) or using big data analysis and machine learning algorithms (Saharuddin et al., 2020; Hosseini et al., 2021). All the approaches reveal that real rubbers and elastomers have to be characterized over a very wide range of frequencies which typically spans many orders of magnitude of the exciting frequency (broad-band behavior), and this, in turn, plays a crucial role in determining the bulk dissipation, hence the interfacial adherence force.

Here we shall consider the problem of a rigid sphere of radius R that is unloaded from a fully relaxed broad-band viscoelastic adhesive half-space (see Fig. 1) presenting and comparing numerical, analytical and experimental results. A few recent works (Violano et al., 2021, 2022; Afferrante and Violano, 2022) have focused on the problem of the adhesion of a rigid Hertzian indenter unloaded from a viscoelastic substrate describing the material either by using (i) the classical three-element solid, also known as the Standard Linear Solid “SLS” (a spring in series with an element constituted by a dashpot and a spring in parallel), (Müser and Persson, 2022; Violano et al., 2022; Afferrante and Violano, 2022) or (ii) by considering the measured response spectrum of the viscoelastic material used in the experimental campaign (Violano et al., 2021, 2022; VanDonselaar et al., 2023). The limitation of the first approach is that the SLS has a narrow-band behavior, hence, although providing valuable insights, it will be rarely useful for modeling the behavior of a real material. The limitation of the second approach is that the results obtained solve only the specific problem considered and it is difficult to draw general conclusions.

The objectives of this work are: (i) to define a material model which may be effectively and efficiently used for describing a real material viscoelastic behavior with a minimal number of constants in both the time and frequency domain, (ii) to determine, for the case of rigid Hertzian indenter-viscoelastic halfspace contact (see Fig. 1), how the broad-band material behavior influences the maximum adherence force as a function of the unloading rate (which is not the speed of the contact radius change, as we shall see) and of the preload, (iii) to provide closed form results for the effective surface energy $\Delta\gamma_{eff}$ based on the Persson and Brener (2005) theory which allows to faithfully reproduce the numerical results together with their region of validity, (iv) to validate the proposed approach by comparing the numerical results with experimental data.

The remainder of the paper is structured to address each objective outlined earlier: Section 2 provides a detailed description of the modified power law model used to characterize the viscoelastic material response; Section 3 presents, the Boundary Element Model developed for analyzing the adhesive contact problem along with extensive numerical results; Section 5 introduces the developed analytical solution and demonstrates its applicability in characterizing adhesive contact problems; Section 6 focuses on the validation of the numerical results based on experimental outcomes; Section 7 discusses the presented results in light of the recent Literature; the manuscript closes with the “Conclusions”, Section 8.

2. Modified power law model

The challenge behind the mechanical modeling of viscoelastic materials arises because the mechanical response at time t depends on the contact history, so that the stress $\sigma(t)$ and strain $\varepsilon(t)$ should be determined by superposition:

$$\varepsilon(t) = \sigma(0)C(t) + \int_0^t C(t-\tau) \frac{d\sigma(\tau)}{d\tau} d\tau, \quad (1)$$

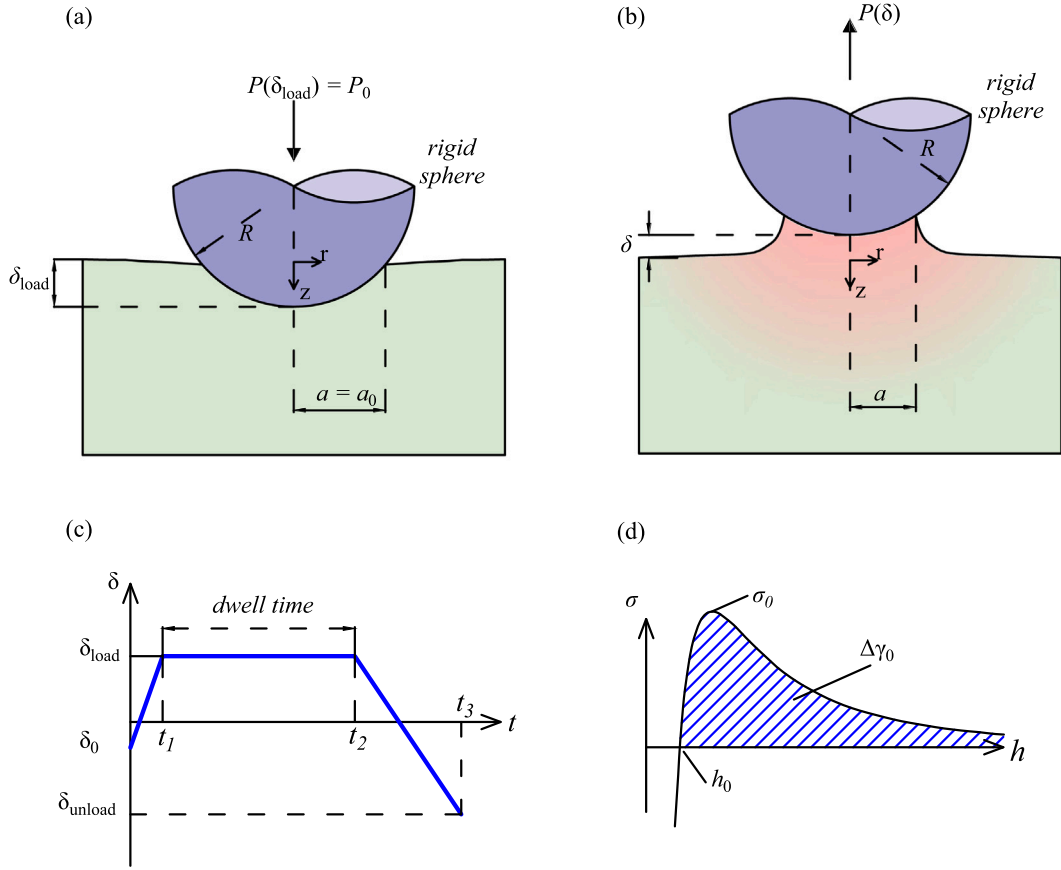


Fig. 1. Sketch of the geometrical model: (a) indentation phase, (b) unloading phase with a constant unloading rate; (c) loading protocol consisting of pre-loading, dwelling, and unloading; (d) the Lennard-Jones force-separation law used at the interface.

$$\sigma(t) = \varepsilon(0) R(t) + \int_0^t R(t-\tau) \frac{d\varepsilon(\tau)}{d\tau} d\tau, \tag{2}$$

where $C(t)$ is the creep compliance function, giving the strain response to a unit stress increment $\sigma(t)$ in uniaxial loading conditions, and $R(t)$ is the relaxation function giving the stress response to a unit strain increment $\varepsilon(t)$ in uniaxial loading conditions. Alternatively, viscoelastic materials can be characterized in the frequency domain. If a sinusoidal stress $\sigma(\omega)$ at frequency ω is applied to a viscoelastic specimen the resulting harmonic strain $\varepsilon(\omega)$ will be delayed by a certain amount δ , hence the so-called complex modulus $\bar{E}(\omega) = \sigma(\omega)/\varepsilon(\omega)$ can be defined in the complex plane. Alternatively, in place of $\bar{E}(\omega)$ one may define its reciprocal, the $\bar{C}(\omega) = \varepsilon(\omega)/\sigma(\omega)$ which is the complex compliance.

One approach to reproduce the broad-band response spectrum of a real viscoelastic material (VanDonselaar et al., 2023; Lorenz et al., 2013) is to move from a SLS material model, which is constituted by a spring in parallel with a single Maxwell element (a spring in series with a dashpot), to the so-called Wiechert model constituted by a spring in parallel with many Maxwell elements (Christensen, 2012), so that several relaxation times can be included in the material representation. Very often the number of elements needed for a faithful representation gets large enough so that the model returns a very good representation of the material viscoelastic behavior, but at the same time it makes it difficult to extract general conclusions, due to the large number of fitting parameters determined.

One option to overcome this difficulty is to rely on power law material models (Schapery, 1975a; Persson and Brener, 2005; Popov et al., 2010; Bonfanti et al., 2020; Dusane et al., 2023), which assume a certain power law function for the distribution of the relaxation times. For example Popov et al. (2010) proposes a model that is fully defined by 5 constants: the relaxed and glassy moduli, two characteristic times, and one exponent. Schapery (1975a) uses an approximation for the creep compliance function $C(t) = (M_e + M_1 t^{-p})^{-1}$ which includes only three material constants $\{M_e, M_1, p\}$ and can describe well the behavior for very long times while being less accurate in describing the short-time material behavior. Furthermore, Persson and Brener (2005) consider a model where the retardation times are distributed as a power law in between two characteristic times and vanishes elsewhere.

In the following, we will consider and extend the Modified Power Law (MPL) material model introduced by Williams (1964), which is fully defined by a minimal set of four parameters: the glassy E_∞ and the rubbery E_0 moduli, a single characteristic time τ_0 and one exponent n . Closed-form results in both time and frequency domains that can be readily used for real viscoelastic material

characterization or as input in viscoelastic crack propagation theories are provided in [Appendix A](#), while in the following the main results are reported.

For the MPL material, the following relaxation spectrum $H(\tau)$ is assumed,

$$H(\tau) = \left(\frac{E_\infty - E_0}{\Gamma(n)} \right) \left(\frac{\tau_0}{\tau} \right)^n \exp\left(-\frac{\tau_0}{\tau}\right), \quad (3)$$

where $\{\tau_0, n\}$ are constants to be determined and $\Gamma(n)$ is the Gamma function. The substitution of Eq. (3) into Eq. (A.3) gives the complex modulus $\bar{E}(\omega) = E'(\omega) + iE''(\omega)$:

$$\bar{E}(\omega) = E_0 + (E_\infty - E_0) i\omega\tau_0 \exp(i\omega\tau_0) \mathbf{E}_n(i\omega\tau_0), \quad (4)$$

where $\mathbf{E}_n(x)$ is the exponential integral function of order $n > 0$. [Appendix A](#) provides closed-form results for both the real $E'(\omega)$ and the imaginary parts $E''(\omega)$ of the complex modulus.

The relaxation function $R(t)$ is given by [Williams \(1964\)](#):

$$R(t) = E_0 + \int_0^\infty \tau^{-1} H(\tau) \exp(-t/\tau) d\tau, \quad (5)$$

which, upon substitution of Eq. (3) gives a very simple form:

$$R(t) = E_0 + \frac{E_\infty - E_0}{(1 + t/\tau_0)^n} \quad (6)$$

or in dimensionless form

$$\hat{R}(\hat{t}) = 1 + \frac{1/k - 1}{(1 + \hat{t})^n} \quad (7)$$

being $\hat{R} = R(t)/E_0$, $\hat{t} = t/\tau_0$ and $k = E_0/E_\infty$, which shows that at a given dimensionless time \hat{t} the material relaxation depends only on the parameters $\{n, k\}$.

Similarly, we can assume a modified power law distribution for the retardation spectrum, as:

$$L(\tau) = \left(\frac{C_0 - C_\infty}{\Gamma(n)} \right) \left(\frac{\tau}{\tau_0} \right)^n \exp\left(-\frac{\tau}{\tau_0}\right), \quad (8)$$

where $\{\tau_0, n\}$ are constants to be determined. Hence the complex compliance is (substitute Eq. (8) into Eq. (A.13)):

$$\bar{C}(\omega) = C_\infty + \frac{(C_0 - C_\infty)}{i\omega\tau_0} \exp\left(-\frac{i}{\omega\tau_0}\right) \mathbf{E}_n\left(-\frac{i}{\omega\tau_0}\right), \quad (9)$$

where $\mathbf{E}_n(x)$ is the exponential integral function of order $n > 0$, $C_0 = 1/E_0$ is the creep compliance in the rubbery limit and $C_\infty = 1/E_\infty$ is the creep compliance in the glassy limit. Notice that once $\bar{C}(\omega)$ is obtained, the complex modulus is also obtained as $\bar{E}(\omega) = 1/\bar{C}(\omega)$ and vice-versa. The [Appendix A](#) reports closed form results for both the real $C'(\omega)$ and the imaginary part $C''(\omega)$ of $\bar{C}(\omega)$.

The creep compliance function $C(t)$ is given by ([Williams, 1964](#)):

$$C(t) = C_\infty + \int_0^\infty \tau^{-1} L(\tau) (1 - \exp(-t/\tau)) d\tau, \quad (10)$$

which, upon substitution of Eq. (8) into Eq. (10) gives:

$$C(t) = C_0 - 2 \frac{(C_0 - C_\infty)}{\Gamma(n)} \left(\frac{t}{\tau_0} \right)^{n/2} \mathbf{K}_n \left(2\sqrt{\frac{t}{\tau_0}} \right), \quad (11)$$

where $\mathbf{K}_n(x)$ is the modified Bessel function of the second kind. The dimensionless creep compliance function $\hat{C} = C/C_0$ is

$$\hat{C}(\hat{t}) = 1 - 2 \frac{(1-k)}{\Gamma(n)} \hat{t}^{n/2} \mathbf{K}_n \left(2\sqrt{\hat{t}} \right), \quad (12)$$

which shows that at a given dimensionless time \hat{t} the material creep depends only on the parameters $\{n, k\}$.

In [Section 6](#), we will show that Polydimethylsiloxane (PDMS, 10:1 resin to curing agent ratio) one of the most common silicone-based polymers used in soft contact mechanics ([Shintake et al., 2018](#); [Sahli et al., 2019](#); [Oliver et al., 2023](#)) has a characteristic exponent at room temperature of $n \simeq 0.22$, which is close to what [Williams \(1964\)](#) found for unfilled HC rubber. In [Fig. 2](#), we illustrate the time evolution of the relaxation and creep compliance functions of MPL for different exponents n (solid black lines) alongside a comparison of the SLS viscoelastic behavior (blue dashed lines). [Fig. 2](#) shows that in order to obtain a behavior close to a standard material, we should set $n \approx 1.6$, which implies PDMS has a much broader spectrum with respect to the SLS. It is recalled that for a SLS the dimensionless creep compliance function is $\hat{C}(\hat{t}) = [1 + (k - 1) \exp(-\hat{t})]$.

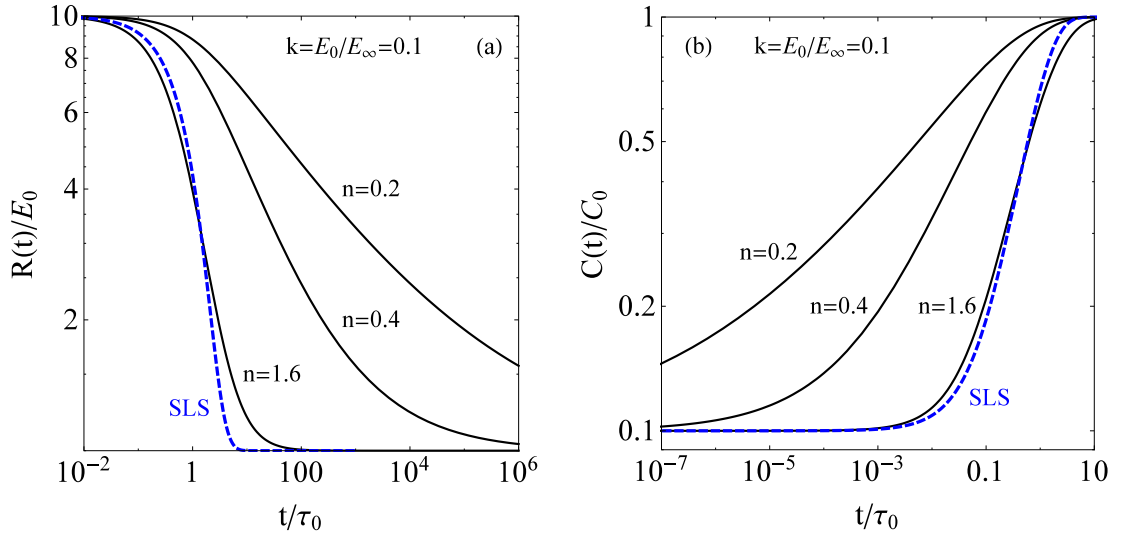


Fig. 2. Time evolution of (a) the relaxation function $R(t)$ (Eq. (6)), (b) the creep compliance function $C(t)$ (Eq. (11)) for $n = [0.2, 0.4, 1.6]$ (solid black line) and a comparison with the behavior of a SLS viscoelastic material (dashed blue line).

3. The numerical model

Let us consider the problem of a rigid sphere of radius R that is unloaded from a fully relaxed viscoelastic adhesive half-space (see Fig. 1). To model the adhesive contact problem a numerical scheme based on the Boundary Element Method was implemented in the software MATLAB, together with a time marching algorithm, which follows the implementation by Papangelo and Ciavarella (2020, 2023). In the numerical model, it is assumed that the interaction between the sphere and the substrate is governed by a Lennard-Jones force-separation law¹:

$$\sigma(h) = -\frac{8\Delta\gamma_0}{3h_0} \left[\left(\frac{h_0}{h}\right)^3 - \left(\frac{h_0}{h}\right)^9 \right], \tag{13}$$

where σ is the interfacial stress ($\sigma > 0$, when compressive), h the local gap, h_0 the equilibrium distance with the surface energy $\Delta\gamma_0 = \frac{9\sqrt{3}}{16}\sigma_0 h_0$. The gap function is then written as:

$$h(r, t) = -\delta + h_0 + \frac{r^2}{2R} + u_z(r, t), \tag{14}$$

where $\delta > 0$ when the sphere approaches the viscoelastic half-space, the sphere profile is approximated by a parabola, and $u_z(r, t)$ is the deflection of the viscoelastic half-space, which depends on the loading history (we have explicitly shown the dependence of u_z on time t). The vertical deflections of the halfspace for an elastic axisymmetric problem are obtained as (Greenwood, 1997; Feng, 2000):

$$u_z(r) = \frac{1}{E_0^*} \int \sigma(s) G(r, s) s ds, \tag{15}$$

where $G(r, s)$ is the Kernel function:

$$G(r, s) = \begin{cases} \frac{4}{\pi r} K\left(\frac{s}{r}\right), & s < r \\ \frac{4}{\pi s} K\left(\frac{r}{s}\right), & s > r \end{cases} \tag{16}$$

$K(k)$ is the complete elliptic integral of the first kind of modulus k and $E_0^* = E_0/(1-\nu^2)$ is the composite elastic modulus (the sphere is rigid) being ν the Poisson ratio considered independent on the excitation frequency ω . Hence, according to the elastic-viscoelastic correspondence principle in the form of Boltzmann integrals (Christensen, 2012), the normal displacements of the viscoelastic half-space $u_z(r, t)$ at time t , at position r will depend on the contact history as:

$$u_z(r, t) = \int G(r, s) s \int_{-\infty}^t C(t-\tau) \frac{d\sigma(s, \tau)}{d\tau} d\tau ds. \tag{17}$$

¹ Strictly speaking Eq. (13) would hold for infinite parallel planes, nevertheless in adhesive contact mechanics it is often assumed that Eq. (13) holds also for slightly inclined surfaces, which is the so-called ‘‘Derjaguin approximation’’, see Greenwood (1997).

where the moduli in the creep compliance function $C(t)$ should be consider in the plane strain conditions, i.e. $C_0^* = 1/E_0^*$. The gap function Eq. (14) is solved through a Newton–Raphson scheme on $N = M + 1$ equally-spaced nodes, being M the number of interfacial elements so that Eqs. (13), (14) and (17) are satisfied. To determine the half-space deflections Eq. (17) was discretized in time and space. In time, we used a time marching algorithm with a time step Δt . In space, we assumed the pressure distribution has a triangular shape over each element, i.e. for the element j th the pressure is p_j at $r = r_j$ and falls linearly to 0 at $r = r_{j-1}$ and $r = r_{j+1}$, which is usually referred as the “method of the overlapping triangles” (Johnson, 1987). Further details of the numerical implementation can be found in Papangelo and Ciavarella (2020, 2023).

4. Numerical results

In the rest of the paper, unless differently stated, the numerical results will be presented in dimensionless notation, as follows:

$$\hat{\delta} = \frac{\delta}{(\pi^2 \Delta \gamma_0^2 R / E_0^*)^{1/3}}; \quad \hat{a} = \frac{a}{(\pi R^2 \Delta \gamma_0 / E_0^*)^{1/3}}; \quad \hat{P} = \frac{P}{\pi \Delta \gamma_0 R}, \quad (18)$$

being $\hat{\delta}$ the dimensionless indentation, \hat{a} the dimensionless contact radius, \hat{P} the dimensionless normal load, $\hat{P}_{po} = |\min(\hat{P})|$ the maximum detachment force, i.e. the pull-off force. Unless specified otherwise, our simulations employ $N = 500$ nodes. Let us consider a sphere with an initial contact radius of a_0 is unloaded from a fully relaxed viscoelastic substrate, with various unloading rates r . This unloading process mimics experimental conditions: (i) indenting the viscoelastic substrate to a specified depth (δ_{load}), (ii) allowing dwell time for substrate relaxation, then (iii) unloading at a constant velocity r . The corresponding dimensionless unloading rate is defined as $\hat{r} = r\tau_0/h_0$. Unless differently stated, the results provided in the following will refer to the Tabor parameter (Tabor, 1977) $\mu = \left(\frac{R\Delta\gamma_0^2}{E_0^*h_0^3}\right)^{1/3} = 3.24$ and $k = E_0/E_\infty = 0.1$.

4.1. Dependence of the detachment force upon the loading protocol details

As it was discussed, the unloading rate r has a crucial role in the mechanical response of viscoelastic materials. We examined our model for different unloading rates while Fig. 3(a) reports only four different unloading rates of $\hat{r} = [10^2, 10^{2.5}, 10^3, 10^6]$. The sphere is unloaded from a fully relaxed substrate with the exponent material of $n = 0.8$ and all the unloading curves in Fig. 3 start from the initial contact radius of $\hat{a}_0 = 3.61$. As anticipated in viscoelastic contact problems, the unloading rate significantly affects the unloading trajectory. Fast unloading boosts viscoelastic dissipation at the crack tip which in turn gives a high pull-off load at detachment. Note that the elastic behavior observed in Fig. 3 corresponds to the initial state of the relaxed substrate.

For the same unloading rate $\hat{r} = 10^{2.5}$, and starting from the same initial contact radius $\hat{a}_0 = 3.61$ the unloading trajectory will be influenced by the response spectrum of the material. In particular, by using the MPL formulation for simulating a broad-band material (see Fig. 2), Fig. 3(b) shows the unloading curves for $n = [0.6, 0.8, 1.6]$, showing that for a given unloading rate the pull-off force generally increases by increasing n . As we will show later, this happens because the more narrow-band the material response spectrum is, the faster the theoretical amplification ($\Delta\gamma_{eff}/\Delta\gamma_0 = E_\infty/E_0$) is reached as a function of the retraction rate.

Finally, the maximum adhesion force at detachment is significantly influenced by the preload, as it was discussed in Violano and Afferrante (2022), Afferrante and Violano (2022). Changing the preload will affect the initial contact area, and also the initial indentation prior to unloading. To demonstrate this effect Fig. 3(c) shows the unloading trajectories of three different unloading curves from the same relaxed viscoelastic substrate (here $n = 1.6$ and $\hat{r} = 10^{2.5}$) while using $\hat{a}_0 = [1.71, 2.47, 3.61]$ respectively. The larger the initial contact radius is, the larger the pull-off force will be. Too small initial contact radius will give rise to finite size effects which will limit the possibility of enhancing the pull-off force up to the theoretical limit predicted by viscoelastic crack propagation theories (Persson and Brener, 2005; Schapery, 1975a).

Hence, when looking for the maximum amplification of the pull-off force, care should be taken in selecting a large enough initial contact radius (or preload). Fig. 4 shows the dimensionless pull-off force \hat{P}_{po} as a function of the dimensionless unloading rate $\hat{r} = r\tau_0/h_0$ for a material with $n = 0.8$ and by changing $\hat{a}_0 = [1.94, 2.50, 3.61, 5.28]$. One realizes that while in the very beginning the enhancement curves all look similar, when the unloading rate starts to increase strong differences appear, with the curves referring to the smaller contact radius providing a much smaller enhancement, below the theoretical value even at the highest unloading rate tested.

Although the dependence of the pull-off force on the initial contact radius and on the unloading rate has been discussed for the paradigmatic SLS model (Violano and Afferrante, 2022; Afferrante and Violano, 2022), it has remained unclear the importance of these parameters for real viscoelastic broad-band materials, hence we will address this question in the next subsection.

4.2. Threshold contact radius

Here, the results of a comprehensive numerical campaign specifically designed for providing, at a given unloading rate, the minimal initial contact radius (or preload) needed to maximize adhesion in a rigid sphere/soft substrate contact are provided. Fig. 5(a) shows the pull-off force obtained unloading the substrate at a given unloading rate $\hat{r} = 10^{0.5}$ as a function of the initial contact radius \hat{a}_0 for $n = [0.4, 0.6, 0.8, 1.6]$ (respectively triangle, star, diamond and square markers). The results show that as \hat{a}_0 increases the pull-off force converges to a certain plateau and that in general, at a given \hat{r} , broad-band materials (low n) will need a smaller initial contact radius to reach convergence of the pull-off force. Hence, in experiments, if the maximum adhesion is sought

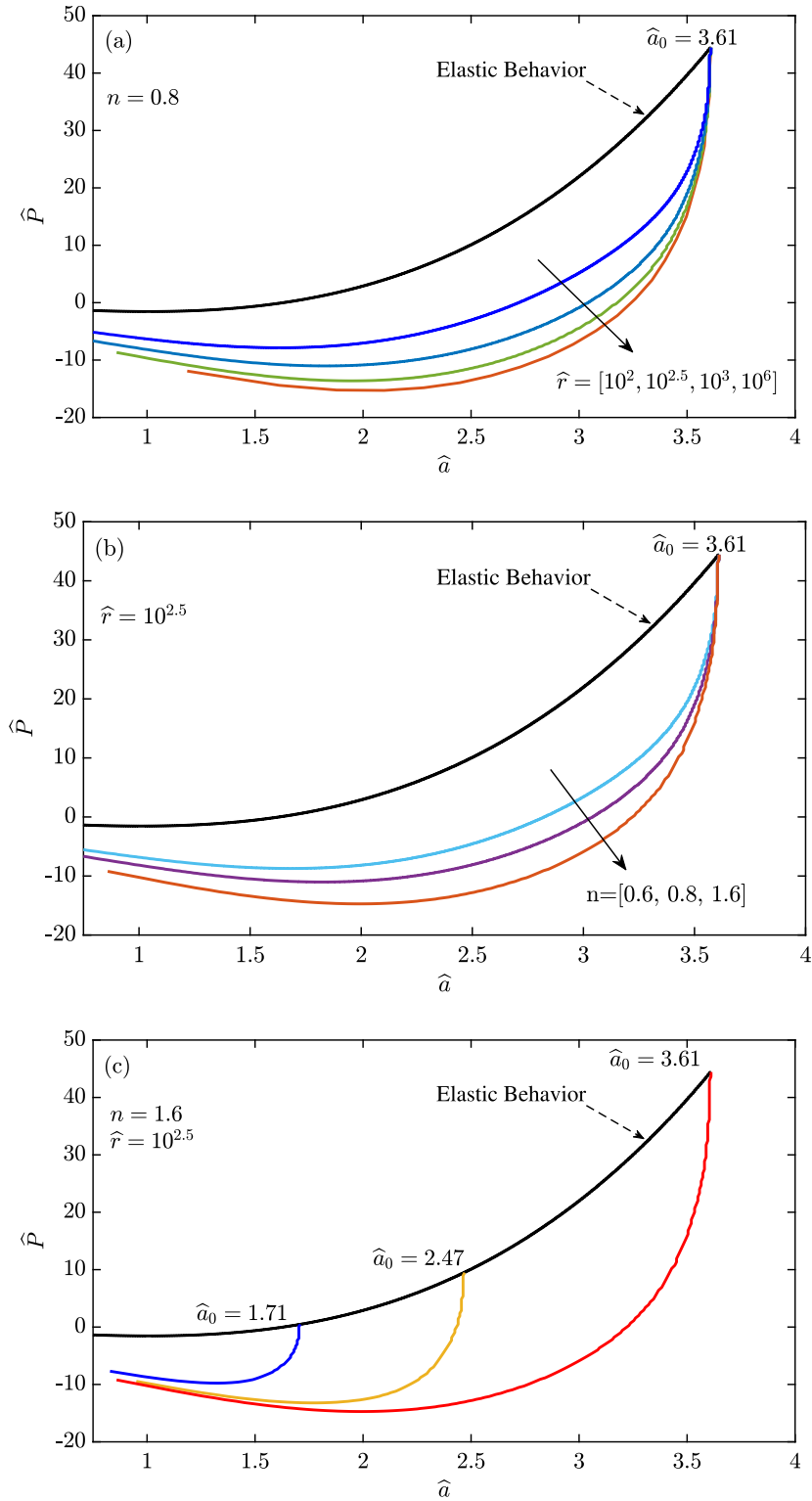


Fig. 3. Dimensionless load \hat{P} versus the dimensionless contact radius \hat{a} . (a) Material exponent $n = 0.8$, initial contact radius $\hat{a}_0 = 3.61$, unloading rates $\hat{r} = [10^2, 10^{2.5}, 10^3, 10^6]$; (b) initial contact radius $\hat{a}_0 = 3.61$, unloading rate of $\hat{r} = 10^{2.5}$ for different material exponents $n = [0.6, 0.8, 1.6]$; (c) initial contact radii $\hat{a}_0 = [1.71, 2.47, 3.61]$, with a constant unloading rate of $\hat{r} = 10^{2.5}$ and material exponent $n = 1.6$. For all the panels unloading starts from a fully relaxed substrate with $k = 0.1$ and $\mu = 3.24$.

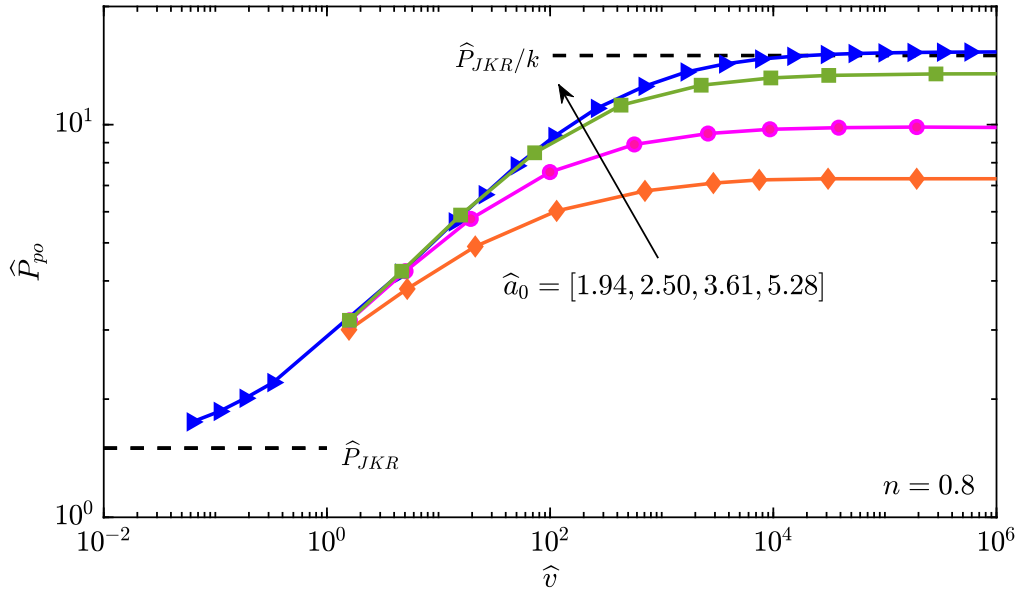


Fig. 4. Normalized pull-off force as a function of the normalized crack velocity for a material with power law exponent $n = 0.8$ and starting the unloading phase from a fully relaxed substrate with initial contact radii $\hat{a}_0 = [1.94, 2.50, 3.61, 5.28]$.

one must first perform a convergence study on the pre-loading conditions. In Fig. 5(a) we have used a spline to interpolate the simulated points (markers), then we have computed the derivative $d\hat{P}_{po}/d\hat{a}_0$ and set the condition $d\hat{P}_{po}/d\hat{a}_0 < 0.1$ to determine a *threshold contact radius* indicated by \hat{a}_{0t} , above which we considered the pull-off force is converged. In Fig. 5(a) the black curves change from dotted to solid when the contact radius is greater than \hat{a}_{0t} .

The results in Fig. 5(a) refer to a particular unloading rate taken as a reference $\hat{r} = 10^{0.5}$. A convergence study was performed over about 5 orders of magnitude in terms of unloading rate, as shown in the inset of Fig. 5(b), where every marker shown corresponds to the threshold contact radius \hat{a}_{0t} , obtained for that material exponent n and at that given normalized unloading rate $\hat{r} \in [10^0, 10^6]$. The inset of Fig. 5(b) explicitly shows a dependence on the viscoelastic material spectrum broadness (i.e. the exponent n). Nevertheless if the data are represented by the normalized pull-off force at convergence as a function of \hat{a}_{0t} they collapse for all the exponents n into a single power law curve that we find to be $\hat{P}_{po} = 0.3\hat{a}_{0t}^{2.7}$ (black dashed line in Fig. 5(b)), which clearly saturates when the maximum enhancement $\hat{P}_{po} = \hat{P}_{JKR}/k = 1.5/k$ is reached. Notice that, the smallest unloading rate considered in our analysis is $\hat{r} \approx 3$ (see Fig. 5(b), inset) as for quasi-static unloading the elastic solution is retrieved and \hat{P}_{po} will not depend on \hat{a}_0 .

Furthermore, Fig. 5(b) shows that the transition from the power-law behavior to the plateau is faster for materials with large material exponent n than for those characterized by low values of n , as a consequence of their narrow spectrum. Hence Fig. 5(b) shows that regardless of the material model, the key parameter that determines the minimum contact radius \hat{a}_{0t} is the maximum amplification of the pull-off force that has to be reached.

5. Persson and Brener crack propagation theory for broad-band viscoelastic materials

In the previous sections, we have shown how the pull-off force depends on the unloading rate and on the initial contact area for various exponent n that characterize the broadness of the viscoelastic material response spectrum. Here, closed-form solutions are obtained for the effective surface energy $\Delta\gamma_{eff}$ based on Persson and Brener (2005) crack propagation theory. It is useful to recall that for a Hertzian indenter, in the case of soft materials, the JKR model (Johnson et al., 1971) applies, which provides the pull-off force depends only on the sphere radius and surface energy $P_{JKR} = \frac{3}{2}\pi R\Delta\gamma_0$, hence, in the following, the normalized effective surface energy will be simply defined as $\hat{\Gamma}_{eff} = \Delta\gamma_{eff}/\Delta\gamma_0 \simeq P_{po}/P_{JKR}$.

We note that our crack propagation formulation is the extension of Persson and Brener (2005) idea of equating the input power from the remote load to the power that is dissipated due to the generation of new surfaces and due to viscoelastic dissipation, so one can obtain the effective surface energy $\Delta\gamma_{eff}$ as (Persson and Brener, 2005):

$$\frac{\Delta\gamma_{eff}}{\Delta\gamma_0} = \left[1 - \left(1 - \frac{E_0}{E_\infty} \right) \int_0^{+\infty} \frac{L(\tau)}{(1/E_0 - 1/E_\infty)\tau} \left\{ \sqrt{1 + b^{-2}(\tau)} - b^{-1}(\tau) \right\} d\tau \right]^{-1}, \quad (19)$$

$$b(\tau) = \frac{2\pi\nu\tau}{l_0} \left(\frac{\Delta\gamma_0}{\Delta\gamma_{eff}} \right). \quad (20)$$

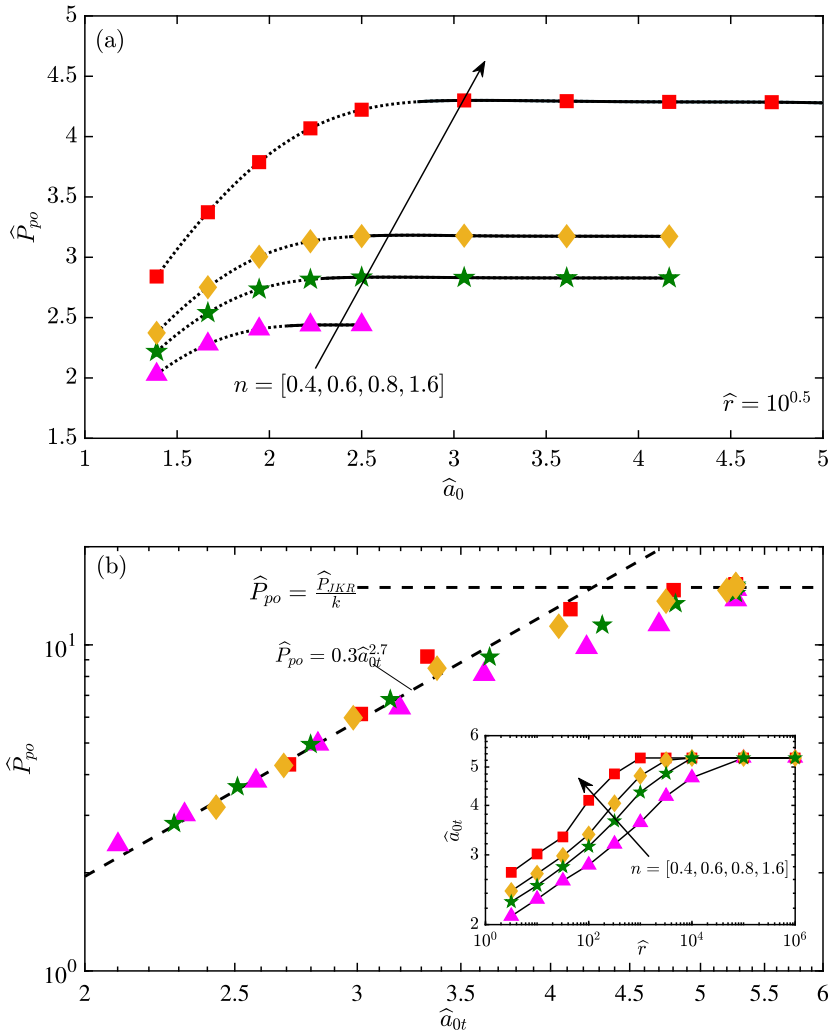


Fig. 5. Threshold contact radius: (a) Dimensionless pull-off force \hat{P}_{po} with respect to the normalized initial contact radius \hat{a}_0 with the same unloading rate of $\hat{r} = 10^{0.5}$ for different material exponents $n = [0.4, 0.6, 0.8, 1.6]$, and $k = 0.1$; (b, inset) Dimensionless threshold contact radius \hat{a}_{0t} with respect to the normalized unloading rate \hat{r} for different material exponents $n = [0.4, 0.6, 0.8, 1.6]$ and $k = 0.1$. (b, main figure) The same data reported in the inset are shown as dimensionless pull-off force \hat{P}_{po} with respect to the normalized threshold contact radius \hat{a}_{0t} . In all the panels triangle, star, diamond and square markers correspond respectively to $n = [0.4, 0.6, 0.8, 1.6]$.

Introducing the dimensionless parameters:

$$\hat{v} = \frac{v\tau_0}{l_0}; \quad \hat{\tau} = \frac{\tau}{\tau_0}, \quad (21)$$

and substituting the retardation spectrum defined for the MPL material model in Eq. (8) into Eq. (19) one gets

$$\hat{\Gamma}_{eff} = \left[1 - (1-k) \int_0^{+\infty} \frac{\hat{\tau}^{n-1} \exp(-\hat{\tau})}{\Gamma(n)} \left[\sqrt{1 + \left(\frac{\hat{\Gamma}_{eff}}{2\pi\hat{v}} \frac{1}{\hat{\tau}} \right)^2} - \left(\frac{\hat{\Gamma}_{eff}}{2\pi\hat{v}} \frac{1}{\hat{\tau}} \right) \right] d\hat{\tau} \right]^{-1}, \quad (22)$$

which can be written as

$$\hat{\Gamma}_{eff} = \left[1 - (1-k) I(n, \hat{v}, \hat{\Gamma}_{eff}) \right]^{-1}, \quad (23)$$

where $I(n, \hat{v}, \hat{\Gamma}_{eff})$ stands for the integral in Eq. (22) which expression is given in closed form in the Appendix B. Following Persson and Brener (2005) original arguments, we determine the lengthscale l_0 equating the linear elastic fracture mechanics stress field to

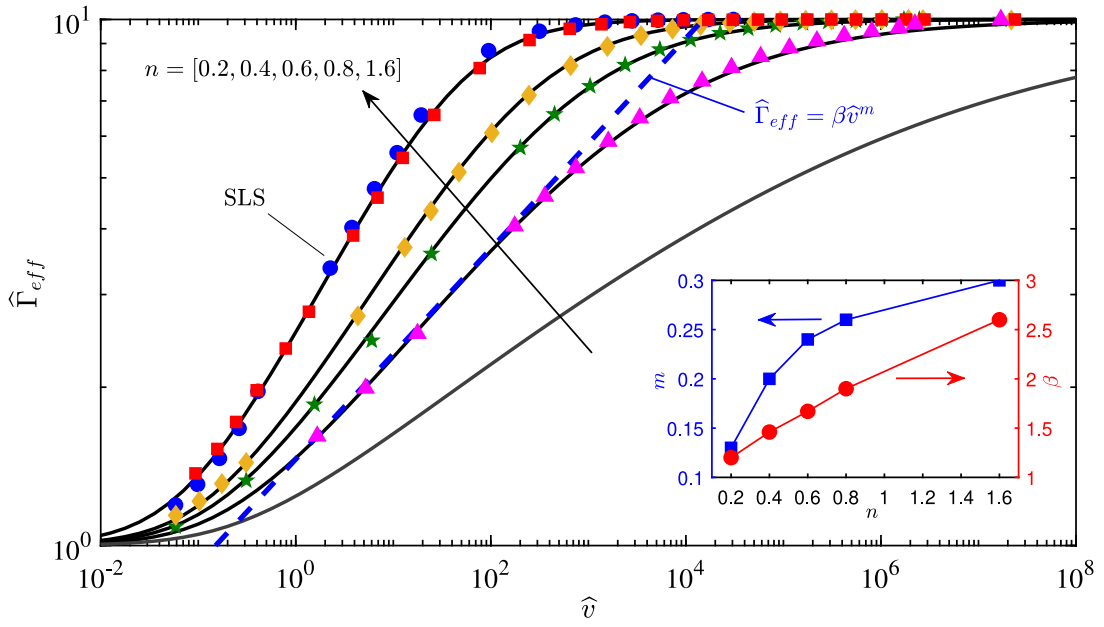


Fig. 6. Normalized effective surface energy $\hat{\Gamma}_{eff}$ with respect to the normalized crack velocity \hat{v} for different power law material exponent $n = [0.2, 0.4, 0.6, 0.8, 1.6]$, respectively triangle, star, diamond and square markers, and $k = 0.1$. The blue circle markers in the plot correspond to the SLS material. Solid lines stand for the PB model (Eq. (23)) for $n = [0.2, 0.4, 0.6, 0.8, 1.6]$. The blue dashed line is a guide to the eye, showing the power law behavior of the function $\hat{\Gamma}_{eff}(\hat{v}) = \beta \hat{v}^m$ in the intermediate velocity range. The inset depicts the fitting parameters $\{\beta, m\}$ for the values of n shown.

the critical stress σ_c required to break the atomic bonds. Hence:

$$\sigma_c = \frac{K_I}{\sqrt{2\pi l_0}}; \quad K_I^2 = 2E_0^* \Delta \gamma_0, \quad (24)$$

$$l_0 = \frac{E_0^* \Delta \gamma_0}{\pi \sigma_c^2} = \frac{E_0^* \Delta \gamma_0}{\pi (\alpha \sigma_0)^2}, \quad (25)$$

where $E_0^* = \frac{E_0}{1-\nu^2}$ is the rubbery plain strain elastic modulus of the halfspace, K_I is the stress intensity factor in mode I and the “2” in its expression takes into account that one of the contacting bodies is rigid, while α in Eq. (25) is a coefficient of order unity to relate the critical stress σ_c in PB theory to the σ_0 we are using in the numerical simulations that are based on the LJ force-separation law. Notice that for soft polymers $l_0/h_0 \approx 1$ hence l_0 should physically be of the same order of the intermolecular distance.

Solving Eq. (23) for $n = [0.2, 0.4, 0.6, 0.8, 1.6]$, $k = 0.1$ and for varying crack velocity \hat{v} one easily find the results shown in Fig. 6 (black solid lines). So, for a given effective energy, broad-band materials would require a much higher crack speed than for narrow-band materials. The numerical results from the same set of parameters are shown in Fig. 6 as markers ($n = [0.4, 0.6, 0.8, 1.6]$), respectively triangle, star, diamond and square markers), where we find an excellent agreement with the analytical results by using $\alpha = \pi/9 \approx 0.3491$. It is reminded that the numerical results shown in Fig. 6 have been obtained unloading a fully relaxed halfspace and are related to an initial contact radius exceeding the threshold value, i.e. $\hat{a}_0 > \hat{a}_{0t}$ (see Fig. 5). Numerical simulations conducted for $k = E_0/E_\infty = [0.01, 0.05, 0.1]$ confirmed that $\alpha \approx \pi/9$ independently on the ratio rubbery to glassy modulus k .

It is worth mentioning that the SLS is very often used as a paradigmatic model for a polymer viscoelastic behavior. As a comparison, Fig. 6 reports the results obtained for the SLS as blue circles, which confirms the case of the SLS is close to $n = 1.6$ and shows a notably large amplification of interfacial adhesion at relatively low crack speed if it is compared with broad-spectrum viscoelastic material. Our experimental results will show in Section 6 that 10:1 PDMS silicone has an exponent $n \approx 0.22$, which implies the maximum adhesion amplification may be observed only at unloading rates which are orders of magnitude larger than that needed for the SLS, which poses also questions about the practical feasibility of reaching so large retraction rates and possible nonlinear effects that may come into play, which will be discussed in the Discussion section. For a more convenient use of Eq. (23), the power law scaling of the effective surface energy in the intermediate velocity range is reported here as $\hat{\Gamma}_{eff} = \beta \hat{v}^m$ (see blue dashed line in Fig. 6), where the parameters β, m can be found in Fig. 6 inset.

The applicability of Eq. (23) for the prediction of the effective surface energy would remain limited by the fact that in all the viscoelastic crack propagation theories, including Eq. (23), the enhancement of the surface energy is a function of the crack velocity at pull-off which is generally not an input parameter in experiments and would be anyway difficult to control. Nevertheless, Fig. 7 shows in the inset that the crack velocity at pull-off \hat{v} scales approximately as:

$$\hat{v} = 2.887 r_{PB}^{1.171}, \quad (26)$$

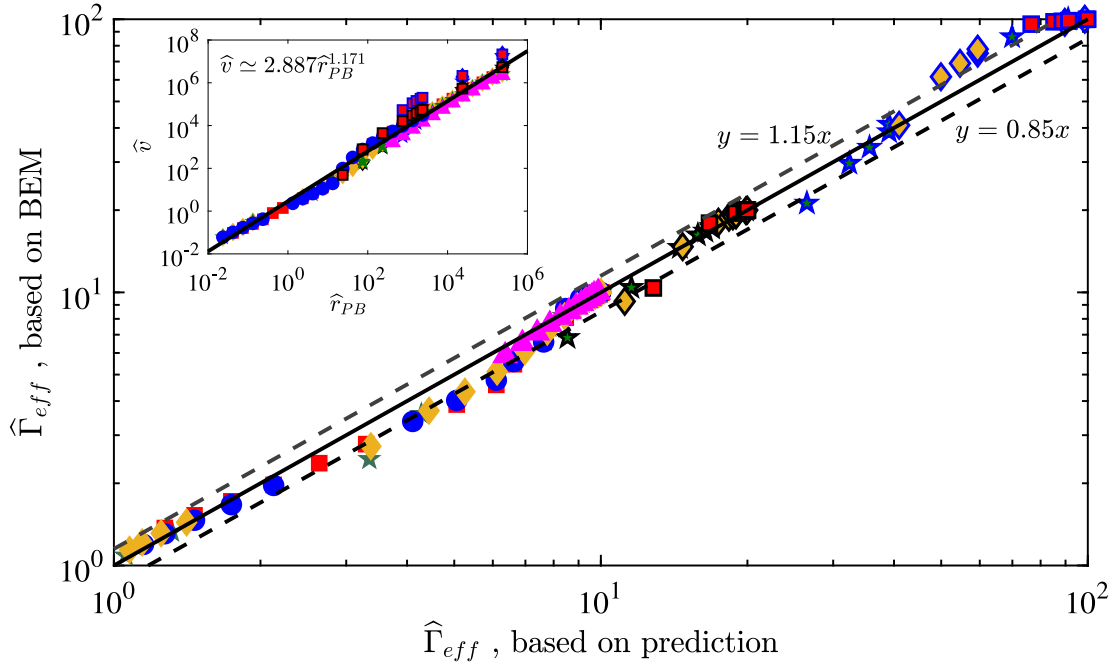


Fig. 7. (main figure) Normalized effective surface energy $\hat{\Gamma}_{eff} = P_{po}/P_{JKR}$ based on the numerical BEM simulations versus the normalized effective surface energy predicted by using the Eqs. (23) and (26). (inset) Normalized crack velocity \hat{v} versus the normalized unloading rate (\hat{r}_{PB}). In both the main figure and the inset the same numerical results are shown, in particular for different power law material exponent $n = [0.4, 0.6, 0.8, 1.6]$, respectively triangle, star, diamond and square markers, and $k = [0.01, 0.05, 0.1]$ respectively markers with a blue contour line, with a black contour line and without contour line. Blue circles stand for the SLS with $k = 0.1$.

over about 10 orders of magnitude in terms of unloading rate \hat{r}_{PB} , where $\hat{r}_{PB} = r\tau_0/l_0$. Fig. 7 shows the numerical results obtained for the material exponents $n = [0.4, 0.6, 0.8, 1.6]$, respectively triangles, stars, diamonds, squares (circles stand for the SLS material) and for $k = [0.01, 0.05, 0.1]$ respectively markers with a blue contour line, with a black contour line and without contour line. Filled blue circles stand for the SLS with $k = 0.1$. Hence, after using Eq. (26) to estimate the crack speed at pull-off as a function of the retraction rate, we have used Eq. (23) to predict the effective surface energy and compared with the numerical BEM results, using the same symbols as in the inset, which are shown in the main Fig. 7. The solid black line represents the condition of the perfect match between prediction and actual numerical results, while as a guide to the eye, we have drawn also two dashed lines representing $\pm 15\%$ error. Although the scaling may be improved by using more refined models, the use of Eqs. (23) and (26) makes the estimate of the pull-off force straightforward based only on the material parameters and on the unloading rate. It is recalled that all the numerical results have been obtained for the Tabor parameter $\mu = 3.24$, hence we expect Eq. (26) to be valid in the limit of short-range adhesion also referred to as the “JKR limit” (Johnson et al., 1971).

6. Experimental adhesion tests

In the previous sections we have developed a general MPL material model capable of describing the viscoelastic behavior of both narrow and broad band materials, then we have compared BEM numerical results with PB theory finding an excellent agreement. Finally, in this section, the numerical predictions will be validated against experimental results.

A series of adhesion tests were performed using a smooth spherical lens loaded and unloaded from a soft viscoelastic substrate at various unloading velocities. The spherical lens was made of borosilicate crown glass (SLB-05-10P, Sigma Koki) with a nominal radius of $R = 5.19$ mm and the substrates were made of polydimethylsiloxane (PDMS, Sylgard 184, DowCorning Corporation) with resin to curing agent weight ratio of 10:1. PDMS is a silicone elastomer well known to exhibit viscoelastic properties, as confirmed in several previous studies (Lorenz et al., 2013; VanDonselaar et al., 2023; Violano et al., 2021; Petroli et al., 2022). For the substrate material characterization, a classical dog-bone shaped specimen was fabricated and used for dynamic mechanical analysis (DMA). All the samples were cured at 70 °C for two hours on a heating table and then followed by natural cooling.

6.1. Material characterization

The DMA test was performed using a DMA850 (TA Instruments) to characterize the viscoelastic properties of the PDMS. The dog-bone-shaped specimen had cross-sectional dimensions of 3.86 mm in width and 0.75 mm in thickness. Temperature sweeps were conducted at a fixed frequency of $f = 1$ Hz and a strain amplitude of $\epsilon = 0.1\%$. The temperature runs from -130 °C to 20 °C with

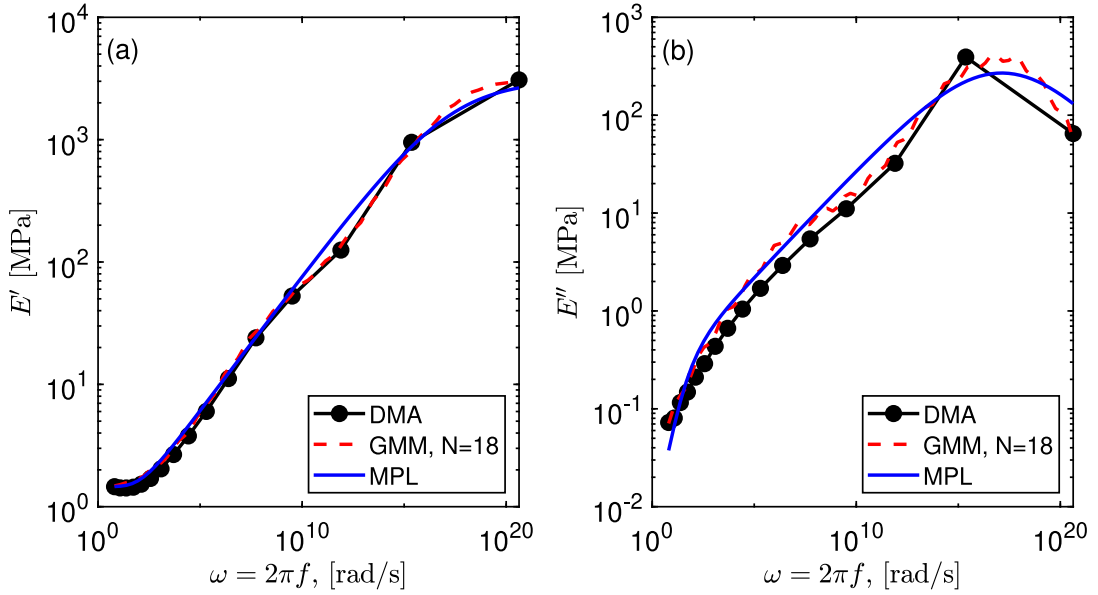


Fig. 8. (a) Real part of the complex elastic modulus E' in the frequency domain at $T_{amb} = 20$ °C. (b) Imaginary part E'' of the complex elastic modulus in the frequency domain at $T_{amb} = 20$ °C. In both panels: the black curve with circle markers stands for the experimental data, the blue curve for the fitted MPL material model and the red curve for the fitted GMM model with 18 arms (see Fig. A.11).

10 °C step size. To move from temperature to frequency domain we used the WLF time–temperature superposition (Williams et al., 1955), hence the shift factor is defined as

$$\log_{10} a_T = \log_{10} \frac{f_{T_g}}{f_T} = \frac{-17.44 (T - T_g)}{51.6 + T - T_g}, \quad (27)$$

where f_T is the frequency at the temperature T and T_g is the glass transition temperature. For the PDMS substrate, we assumed $T_g = -115$ °C, which agrees well with the results reported in Ref. VanDonselaar et al. (2023) for the same material. Furthermore, we note that using $T_g = -115$ °C our measurements of the complex modulus \bar{E} also satisfy the Kramers–Kronig(KK) relation (Pritz, 2005)

$$E''(\omega) = -\frac{2\omega}{\pi} \int_0^{+\infty} \frac{E'(u)}{\omega^2 - u^2} du, \quad (28)$$

where $\omega = 2\pi f$ is the angular frequency and the integral should be intended as its Principal Value (Pritz, 2005).

The experimental data for the complex modulus were shifted to $T_{amb} = 20$ °C by using Eq. (27) and fitted using Eq. (9), which is written in terms of the complex compliance $C(\omega)$ as that is the function needed in the numerical BEM implementation (see Eq. (17)). Fig. 8, panels (a)–(b), shows the complex modulus $\bar{E}(\omega) = 1/\bar{C}(\omega) = E'(\omega) + iE''(\omega)$ as obtained experimentally (black solid curve with circle markers) and as fitted by the MPL material model (blue solid curve). For PDMS we found

$$\begin{aligned} E_0 &= 1.458 \text{ MPa} \\ E_\infty &= 3.089 * 10^3 \text{ MPa} \\ n &= 0.2207 \\ \tau_0 &= 0.01876 \text{ s} \end{aligned} \quad (29)$$

For comparison purposes, the result that would have been obtained by fitting the experimental data using a Generalized Maxwell Model (GMM, also known as the “Wiechert model”) with 18 arms, hence 37 constants, is also shown in Fig. 8 as a dashed red curve. One realizes that both the GMM and the MPL models give a fair representation of the material behavior, although the MPL model is simpler to use, and the four parameters used in the fitting procedure $\{E_0, E_\infty, n, \tau_0\}$ have a straightforward physical interpretation.

6.2. Experimental setup and comparison

A custom-designed adhesion test instrument, based on the tribometer platform (NTR2, CSM Instruments), was constructed to measure the pull-off force. As illustrated in Fig. 9(a), the lens was rigidly fixed to the force sensor. The PDMS substrate was positioned above a transparent rigid box, with the contact interface observable through a camera via a prism mounted inside the box. The pull-off tests comprised three sequential steps: loading, dwelling, and unloading. Initially, the lens was gradually loaded against the PDMS substrate with a preload force denoted as P , followed by a dwell period of 60 s to ensure complete relaxation of adhesive contact.

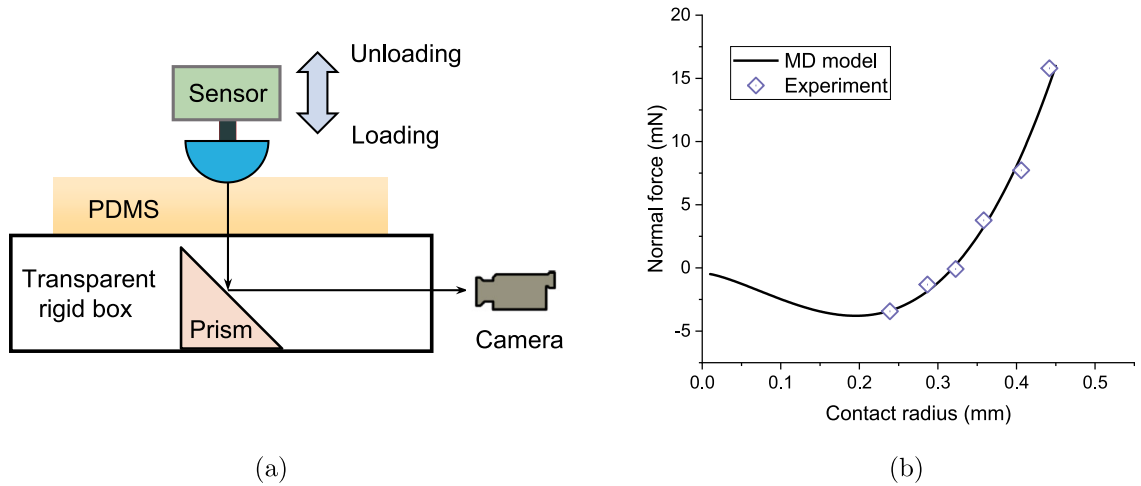


Fig. 9. (a) Schematic of the experimental setup for adhesion tests; (b) Variations of normal force with contact radius a at a very low unloading rate to determine the interfacial parameters.

Subsequently, the lens was pulled out at a fixed unloading rate, r . Throughout the entire process, the normal force was recorded and the pull-off force represents the absolute minimum normal force. Firstly, we measured the variation of the normal force with the contact radius, a , at a very low unloading rate $r = 0.98 \mu\text{m/s}$ to determine the interfacial parameters, as shown in Fig. 9(b). By fitting the relationship between the normal force and contact radius using Carpick's method (Carpick et al., 1999), we estimated the intrinsic work of adhesion $\Delta\gamma_0 = 0.152 \text{ J/m}^2$ and the Tabor parameter $\mu = 2.05$. Next, we conducted tests by varying the unloading rate r . The lens is brought into contact with the PDMS substrate and loaded to the preset preload $P_0 = 1.5 \text{ mN}$. After a 60-second dwell period, the lens is moved upward until the contact is broken and the lens is pulled off from the substrate. We used our numerical BEM code, using the MPL material model for the viscoelastic substrate, to predict the pull-off force during the unloading process. The comparison with experiments leads to the result shown in Fig. 10, where the pull-off force, P_{po} [mN], is plotted as a function of the unloading rate, r [$\mu\text{m/s}$] (the red squares stand for the experimental data, the black solid line for the numerical results). According to Fig. 10, the experimental results confirm an increase in the pull-off force with increasing unloading rates. While there is good agreement between numerical and experimental results in the range of retraction rates $r = [1, 100] \mu\text{m/s}$, the experimental data exhibit marked higher values than the numerical predictions for high values of the unloading rate $r > 100 \mu\text{m/s}$, which agrees well with other published experimental results (VanDonselaar et al., 2023; Tiwari et al., 2017). For having a good fit of the low speed experimental results, we set $h_0 = 30.8 \text{ nm}$, which is discussed in detail in the Discussion section.

7. Discussion

7.1. On the possibility of reaching the maximum adhesion enhancement

According to viscoelastic crack propagation theories (Persson and Brener, 2005; Schapery, 1975a,b) the maximum enhancement of the pull-off force is $\hat{P}_{po} = \hat{P}_{JKR}/k$, hence based on the results reported in Section 4, one can estimate that for a PDMS material with $k \simeq 4.73 \times 10^{-4}$ the maximum amplification of the pull-off force will be reached for $\hat{a}_{0r} \simeq 30.9$. Using the interfacial and material properties we have found for PDMS ($\Delta\gamma_0 = 152.3 \text{ mJ/m}^2$, $\nu = 0.5$, $E_0^* \simeq 1.94 \text{ MPa}$) and for $R = 5.19 \text{ mm}$ gives an initial contact radius of $a_{0r} = 5.8 \text{ mm}$, which is larger than the sphere radius and even considering a parabolic (Hertzian) profile certainly outside the limit of validity of the hypothesis of small deformations, which raises doubts about the practical feasibility of reaching the maximum amplification factor predicted by crack propagation theories.

Another consideration to be made is related to the unloading rate that would be needed to reach the maximum adhesion amplification. By using the results reported in Section 6, one estimates that to reach the maximum amplification for a PDMS substrate one would need to unload the substrate at $\hat{v} \approx 10^9$ (see Fig. 6), which using $l_0 = 30.0 \text{ nm}$ and $\tau_0 = 0.01876 \text{ s}$ together with Eq. (26) gives the dimensional unloading rate of about $r \approx 31.4 \text{ m/s}$, which is about 4 orders of magnitude larger than the maximum unloading velocity usually used in adhesion experiments (Tiwari et al., 2017; VanDonselaar et al., 2023), provided also the limitations introduced by the inertia of the motorized linear stages. Hence, insufficient preload and unloading rates used in experiments may partially explain why, in Literature, measurements of very large enhancement factors, even close to $1/k$, are missing (see for example VanDonselaar et al., 2023; Tiwari et al., 2017).

Our numerical and experimental results seem to be in agreement with the experimental adhesion tests reported in VanDonselaar et al. (2023), Tiwari et al. (2017) for a similar PDMS material, where they also found that PB theory agreed well with experimental observations only up to about $r \approx 100 \mu\text{m/s}$. This may suggest that both numerical and theoretical models are lacking essential phenomena to describe the detachment process at high retraction rates. At present, a few hypotheses have been formulated, ranging

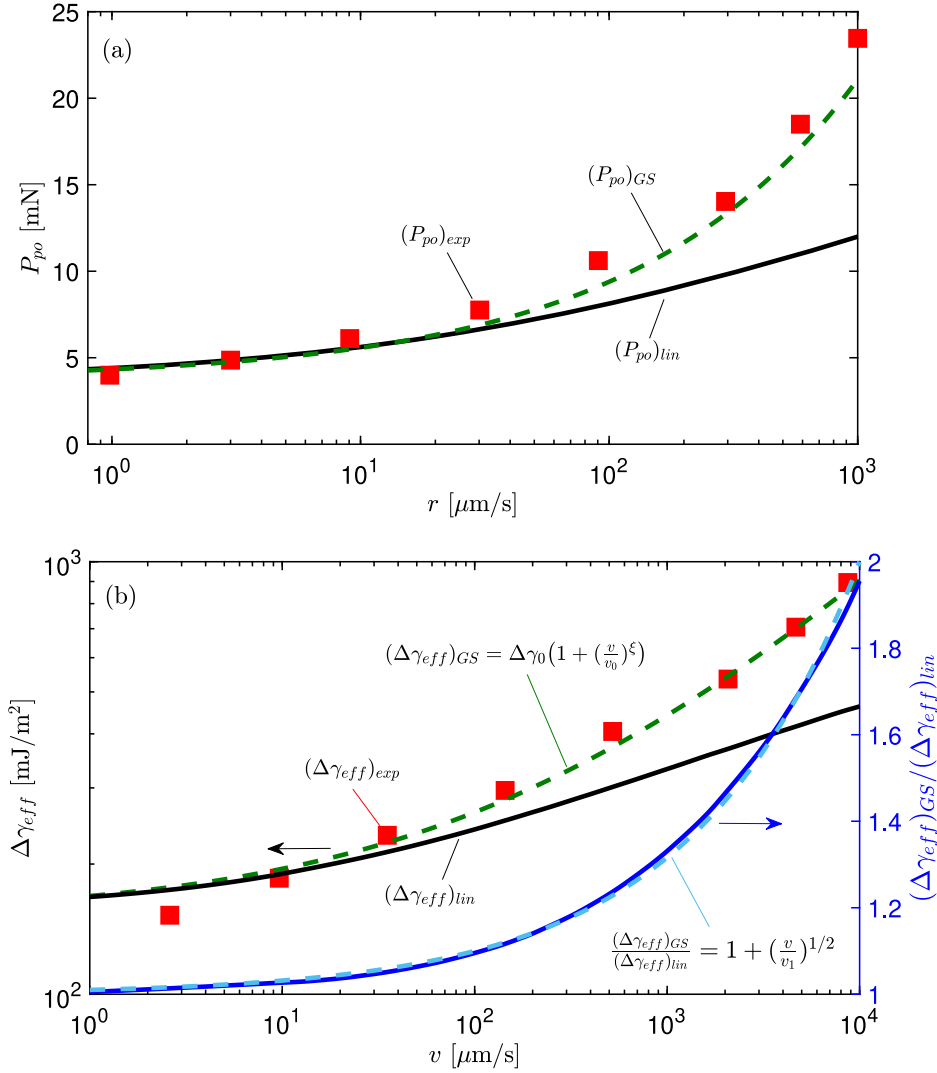


Fig. 10. (a) Pull-off force as a function of the unloading rate: comparison between numerical (from the BEM code, solid black line, labeled $(P_{po})_{lin}$) and experimental results (red square markers) for preload $P_0 = 1.5$ mN, $R = 5.19 \times 10^{-3}$ m, $\Delta\gamma_0 = 152.3$ mJ/m², $h_0 = 30.8$ nm. The green dashed line was obtained using the effective surface energy fitted on the experimental results labeled as $(\Delta\gamma_{eff})_{GS}$ and shown in panel (b). (b, left y-axis) Effective surface energy from: the experimental results (red squares, labeled $(\Delta\gamma_{eff})_{exp}$), the fit of the experimental data using a Gent and Schulz power law model (Eq. (30), where $v_0 = 213.5$ $\mu\text{m/s}$ and $\xi = 0.4154$, dashed green line, labeled $(\Delta\gamma_{eff})_{GS}$), the prediction obtained for the PDMS substrate using the linear numerical BEM model (solid black line, labeled $(\Delta\gamma_{eff})_{lin}$). In this respect we used our approximate Eq. (26) to determine the crack velocity at pull-off starting from the experimental retraction rate. (b, right y-axis) The ratio between $(\Delta\gamma_{eff})_{GS}/(\Delta\gamma_{eff})_{lin}$ for which a power law fit is provided $\frac{(\Delta\gamma_{eff})_{GS}}{(\Delta\gamma_{eff})_{lin}} = 1 + (\frac{v}{v_1})^{1/2}$ where $v_1 = 10^4$ $\mu\text{m/s}$ (dashed pale blue curve).

from the possibility of nonlinear dissipative phenomena, happening within the process zone, like (i) cavitation and stringing, (ii) extraction of non-cross linked polymeric chains from the substrate, (iii) temperature dependence of the material behavior at the crack tip, (iv) the nonlinear behavior of the material at the large strains ($\approx 10\%$) experienced close to the crack tip (VanDonselaar et al., 2023; Tiwari et al., 2017), of course not included into the (linear) theoretical and numerical models, which is discussed in the next subsection.

7.2. Energy dissipation within the process zone

To obtain a satisfactory fit of the experimental data at low unloading rates we set $h_0 = 30.8$ nm. Notice that from quasi-static experiments, using the definition of the Tabor parameter we would have obtained a much larger equilibrium distance $h_0 = 1.55$ μm , which is close to what can be obtained for the same PDMS material using the parameters in Oliver et al. (2023). This calls for further investigations in the future.

For the PDMS material we have characterized, using $h_0 = 30.8$ nm, one obtains that the size of the process zone that fits the experimental data in Fig. 10a is $l_0 \approx 30.0$ nm. Indeed, determining the length of the process zone in viscoelastic crack propagation is still an open question. In the de Gennes (1996) and Saulnier et al. (2004) theories, the size of the “nonlinear” zone is assumed to be a constant, and the fracture energy has its maximum amplification at intermediate speeds. In PB theory this size is not constant and is directly proportional to the applied energy release rate G , which results in a model practically coincident with the cohesive zone model of Knauss and Schapery (see Knauss, 2015). However, in fitting experimental data of fracture (Hui et al., 2022) consider two examples, a styrene-butadiene co-polymer from Gent and Lai (1994), where they do not have independent estimate of the cohesive strength, but simply fit the fracture energy vs speed data, obtaining a process zone size at low speed of a nonphysical size of 0.1 nm, consistent with Gent and Lai (1994). In the second example, they consider a polyurethane elastomer called Solithane 113 of Knauss (2015), and obtain by the same process a size of 1 nm. Hence, Hui et al. (2022) conclude that this size cannot realistically represent a dissipation zone for which a lower bound should be the length of the monomer unit ≈ 46 nm (Lake and Thomas, 1967).² Notice that nonlinear crack propagation theories have been developed by Schapery using cohesive models (Schapery, 1984), and have provided a fracture process zone at low speeds of approximately 10 nm, much more realistic than the 0.1 nm found by Knauss (2015) and Schapery (1975b) for Solithane rubber. In fitting crack propagation data in rubbers, Schapery (2022a) (Tab. 1) found a jump in propagation speed at a certain applied load which seems to suggest a sharp change of cohesive zone fracture energy as a function of speed. He found a low speed fracture energy which is higher than the fast propagation speed fracture energy of a factor of about 6. In our adhesion experiments, our theory is linear and hence we cannot exclude that a nonlinear theory would explain this apparent continuous change of cohesive zone fracture energy with the speed of the linear theory, which is an increase with speed rather than a decrease and hence gives no instability.

Barthel (2024) reports post mortem experimental measurements of the process zone length from damage that occurred at the crack tip and shows this should be of a physically reasonable size of the order of microns. Also, it clearly increases with size as PB and Schapery suggest, but contrary to the original DeGennes and Saulnier theories. Furthermore, linear theories seem to work better for very viscoelastic solids, namely when the glass transition temperature is above ambient temperature, perhaps because for very viscoelastic materials the dissipation in the bulk becomes dominant (Barthel, 2024).

We have estimated the experimental effective surface energy $\Delta\gamma_{eff}(v)$ (Fig. 10b, left y-axis) as obtained from experiments (red squares), fitted by a Gent-Schultz (Gent and Schultz, 1972) power law equation (green dashed curve)

$$\Delta\gamma_{eff} = \Delta\gamma_0 \left(1 + \left(\frac{v}{v_0} \right)^\xi \right), \quad (30)$$

and estimated from our linear BEM numerical scheme (black solid line), respectively $\{(\Delta\gamma_{eff})_{exp}, (\Delta\gamma_{eff})_{GS}, (\Delta\gamma_{eff})_{lin}\}$ in Fig. 10b. To estimate the crack velocity at pull-off from the retraction rates used in the experiments we used the approximate relationship in Eq. (26), and this shows that a linear theory would fit the data much better (see dashed green line in Fig. 10) if we assume a rate-dependent surface energy. Notice that PDMS (10:1) elastic behavior (quasi-static loading, room temperature) has been well characterized and it shows a very neat linear behavior up to $\approx 50\%$ strain (Akogwu et al., 2010), hence, in principle, one would expect PB theory should work, unless other causes of dissipation are at play.

Indeed, even considering that $l_0 \approx 30.0$ nm is a more realistic estimate of the length of the fracture process zone, still we have shown that above $v = 100$ $\mu\text{m/s}$ the linear theory largely underestimates the effective surface energy as shown in Fig. 10b. Hence, other rate-dependent causes of dissipation seem to be at play which consistently contribute to determine the overall energy to be spent for the crack to propagate.

As we have demonstrated numerically, linear theories such as PB theory, successfully estimate the dissipation happening within the bulk material, but they fail to account for the rate-dependent nonlinear dissipative processes taking place within the process zone. Clearly, the assumption of constant intrinsic fracture energy and cohesive stress in the cohesive zone where large strain, high strain rate and nonlinear deformations (including damage) happen, is questionable as noticed by a very recent contribution by Barthel (2024). Introducing the dissipative contribution coming from the nonlinear phenomena happening within the process zone would ultimately require additional constants to be determined from actual measurements, unless one aims at describing all the nonlinear processes happening within the process zone. Unfortunately, the way the effective surface energy $\Delta\gamma_{eff}$ increases with speed would require a separate investigation as this is a completely different contribution than the one coming from viscoelastic bulk dissipation and, at present, the scientific community has not agreed on a good model for it (Bažant and Li, 1997; Lavoie et al., 2015; Musto and Alfano, 2013; Zhao et al., 2021).

Given the considerable effort in the theory in characterizing the viscoelastic linear properties, these recent models are trying mostly to understand how much of the fracture energy amplification comes from the bulk dissipation and how much from the cohesive zone process rate-dependence. In this respect, the estimate we gave in Fig. 10b suggests that in our experiments at $v = 10^4$ $\mu\text{m/s}$ the nonlinear rate-dependent dissipative contribution originated within the process zone $(\Delta\gamma_{eff})_{GS} - (\Delta\gamma_{eff})_{lin}$ equals the one coming from the dissipation in the bulk $(\Delta\gamma_{eff})_{lin}$ (blue curve).

² Recent literature contributions have started to question the validity of classical linear elastic fracture mechanics for unfilled plastics and elastomers suggesting that fracture initiates at a critical tensile strength, see Wang et al. (2024).

8. Conclusions

We have studied the adhesive contact between a rigid Hertzian indenter and a substrate constituted by a broad spectrum viscoelastic halfspace. For the material we have adopted a Modified Power-Law (MPL) material model, originally proposed by Williams (1964), that we have extended to provide closed-form results for the creep compliance function and for the relaxation function in the time domain, and also for the complex modulus and the complex compliance in the frequency domain. Notably, the MPL model is a function of only 4 parameters, the two moduli, a characteristic exponent n and a characteristic time τ_0 . In particular, by changing the exponent n , we have shown that it is possible to have a realistic description of a broad-band viscoelastic material, which we have demonstrated by fitting the complex modulus measured for a PDMS sample.

By using a numerical model based on the Boundary Element Method (BEM), extensive numerical studies have been performed in a wide range of the unloading rate, spanning about 8 orders of magnitude. We have shown that due to viscoelasticity, the effective surface energy can be strongly enhanced with respect to the thermodynamic surface energy, nevertheless to avoid finite size effects a certain minimum contact radius has to be reached, which we named a “threshold contact radius” a_{0t} . Our numerical simulations have shown that a_{0t} is independent of the material exponent, but it depends on the pull-off enhancement that has to be reached at high unloading velocity.

Provided that finite size effects are avoided ($a_0 > a_{0t}$), the theory of Persson and Brener (2005) can be used to determine the pull-off force of the spherical indenter as a function of the crack speed at pull-off with high accuracy, but only within the assumptions of the linear theory and rate-independent fracture process zone parameters. Relating the numerical results based on a Lennard-Jones force-separation law to the theory of Persson and Brener (2005) required to define a parameter $\alpha = 0.3491$ of order unity that relates the critical stress σ_c in PB theory to the maximum stress used in the LJ law σ_0 , which was found independent on the ratio $k = E_0/E_\infty$. Adhesion experiments are usually run in displacement control, and the crack speed at pull-off is certainly not a control parameter, nevertheless, we have shown that the velocity of the crack at pull-off \hat{v} scales as $\hat{v} \simeq 2.887\hat{r}_{PB}^{1.171}$ over more than 8 orders of magnitude, which provides an extremely simple relation to roughly estimate the pull-off force starting only from the material model parameters and the unloading rate with about $\pm 15\%$ confidence.

Finally, by using the MPL for the viscoelastic material and the developed BEM code, we have attempted a comparison between the numerical and the experimental results, which turned out to be satisfactorily accurate up to unloading rates $r = 100 \mu\text{m/s}$, while for faster unloading the numerical results predict lower enhancement with respect to what is measured by our experiments. This observation turns out to be in good agreement with previous Literature results VanDonselaar et al. (2023), Tiwari et al. (2017), where similar experiments were conducted.

A nonlinear description of the material behavior must be necessarily a better description than linear, so perhaps the J integral approach of Schapery (Schapery, 2023) could improve our results. However, as in classical nonlinear fracture mechanics, we ultimately need to measure experimentally the critical value of the fracture energy, which cannot be found reliably from other material properties, in viscoelastic adhesion even if some progress is made by the linear theories, the estimate of the bulk dissipation contribution to fracture energy enhancement is not sufficient, and, ultimately, the fracture process zone rate-dependency must be measured experimentally. Hence, at present, the measurement of the $\Delta\gamma_{eff}(v)$ curve remains the only engineering approach, resulting in the phenomenological (Gent and Schultz, 1972) law. Notice that if we use the measured Gent-Schultz law with a power $\xi = 0.41$ and assume the far field material is elastic with relaxed modulus, we can solve the adhesive contact problem using the Muller solution as corrected in Ciavarella (2021). This results in a pull-off force that does not scale with the same power law of the Gent-Schultz law, but with power 0.27 in this case, so also the Muller solution is misleading.

CRediT authorship contribution statement

Ali Maghami: Writing – original draft, Software, Investigation, Data curation. **Qingao Wang:** Writing – original draft, Methodology, Investigation, Data curation. **Michele Tricarico:** Supervision, Investigation. **Michele Ciavarella:** Writing – review & editing, Writing – original draft, Validation, Supervision, Methodology, Investigation, Conceptualization. **Qunyang Li:** Writing – review & editing, Supervision, Resources, Funding acquisition. **Antonio Papangelo:** Writing – review & editing, Writing – original draft, Supervision, Resources, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The dataset generated for this article is available on Zenodo at <https://doi.org/10.5281/zenodo.13358696>.

Acknowledgments

A.M., M.T., M.C. and A.P. were partly supported by the Italian Ministry of University and Research under the Programme “Department of Excellence” Legge 232/2016 (Grant No. CUP - D93C23000100001). A.P., A.M. and M.T. were supported by

the European Union (ERC-2021-STG, “Towards Future Interfaces With Tuneable Adhesion By Dynamic Excitation” - SURFACE, Project ID: 101039198, CUP: D95F22000430006). Views and opinions expressed are however those of the authors only and do not necessarily reflect those of the European Union or the European Research Council. Neither the European Union nor the granting authority can be held responsible for them. A.P. was partly supported by the European Union through the program – Next Generation EU (PRIN-2022-PNRR, “Fighting blindness with two photon polymerization of wet adhesive, biomimetic scaffolds for neurosensory REtina-retinal Pigment epitheliAl Interface Regeneration” - REPAIR, Project ID: P2022TTZZF, CUP: D53D23018570001). Q.W. was supported by National Natural Science Foundation of China (No. 12025203).

Appendix A. Modified power law material model

A.1. Relaxation function in time and frequency domain

Let us assume to model a viscoelastic material with a continuous distribution $H(\tau)$ of relaxation times, which is the so-called material relaxation spectrum, in parallel with a Hookean spring giving the material stiffness for a long time. This coincides with assuming a Wiechert model (see Fig. A.11) with an infinite number of Maxwell arms. The general relation for the stress $\sigma(t)$ at time t is (Eq. (2.34) in Williams (1964))³:

$$\sigma(t) = \left\{ E_0 + \int_0^\infty \frac{H(\tau)}{\left[\frac{d}{dt} + 1/\tau\right]\tau} d\tau \frac{d}{dt} \right\} \varepsilon(t), \tag{A.1}$$

Converting Eq. (A.1) in the frequency domain, we get:

$$\sigma(\omega) = \left\{ E_0 + \int_0^\infty \frac{H(\tau) i\omega}{[i\omega\tau + 1]} d\tau \right\} \varepsilon(\omega) = \bar{E}(\omega) \varepsilon(\omega), \tag{A.2}$$

where, i is the imaginary unit, ω is the angular frequency, and, by definition, $\bar{E}(\omega) = E'(\omega) + iE''(\omega)$ is the complex modulus, hence:

$$\bar{E}(\omega) = E_0 + \int_0^\infty \frac{H(\tau) i\omega}{[i\omega\tau + 1]} d\tau. \tag{A.3}$$

In order to fit the experimental data, one can guess a certain form for the relaxation spectrum $H(\tau)$. As suggested by Williams (1964), a broad-band approximation of the response of the viscoelastic material can be obtained by adopting for the relaxation spectrum a modified power law:

$$H(\tau) = \left(\frac{E_\infty - E_0}{\Gamma(n)} \right) \left(\frac{\tau_0}{\tau} \right)^n \exp\left(-\frac{\tau_0}{\tau}\right), \tag{A.4}$$

The complex modulus is $\bar{E}(\omega) = E'(\omega) + iE''(\omega)$ can be written in terms of the relaxation spectrum:

$$\bar{E}(\omega) = E_0 + \int_0^\infty \frac{H(\tau) i\omega}{[i\omega\tau + 1]} d\tau, \tag{A.5}$$

$$E'(\omega) = E_0 + \int_0^\infty \frac{H(\tau) \omega^2 \tau}{[1 + \omega^2 \tau^2]} d\tau, \tag{A.6}$$

$$E''(\omega) = \int_0^\infty \frac{H(\tau) \omega}{[1 + \omega^2 \tau^2]} d\tau. \tag{A.7}$$

By using Eq. (A.4) for the relaxation spectrum $H(\tau)$ one obtains

$$\bar{E}(\omega) = E_0 + (E_\infty - E_0) i\omega\tau_0 \exp(i\omega\tau_0) \mathbf{E}_n(i\omega\tau_0), \tag{A.8}$$

$$E'(\omega) = E_0 + \frac{(E_\infty - E_0)}{\Gamma(n)} \left\{ \begin{aligned} &\pi (\tau_0\omega)^n \cos\left(n\frac{\pi}{2} + \tau_0\omega\right) \csc(n\pi) + \dots \\ &\dots + (\tau_0\omega)^2 \Gamma(n-2)_p \mathbf{F}_q \left[1; \left\{ \frac{3-n}{2}, 2 - \frac{n}{2} \right\}; -\frac{(\tau_0\omega)^2}{4} \right] \end{aligned} \right\}, \tag{A.9}$$

$$E''(\omega) = \frac{(E_\infty - E_0)}{\Gamma(n)} \left\{ \begin{aligned} &\pi (\tau_0\omega)^n \sin\left(n\frac{\pi}{2} + \tau_0\omega\right) \csc(n\pi) + \dots \\ &\dots + (\tau_0\omega) \Gamma(n-1)_p \mathbf{F}_q \left[1; \left\{ 1 - \frac{n}{2}, \frac{3-n}{2} \right\}; -\frac{(\tau_0\omega)^2}{4} \right] \end{aligned} \right\}, \tag{A.10}$$

where τ_0 is the characteristic time, $n > 0$ is a characteristic exponent, ω is the angular frequency, E_0 is the relaxed elastic modulus, E_∞ is the instantaneous elastic modulus, ${}_p\mathbf{F}_q[a; b; z]$ is the generalized hypergeometric function, $\Gamma(x)$ is the Euler gamma function.

³ Notice that we are using the notation according to Williams (1964). In Christensen (2012) book the relaxation spectrum is defined as $[H(\tau)]_{Christensen} = [H(\tau)]_{Williams} / \tau$

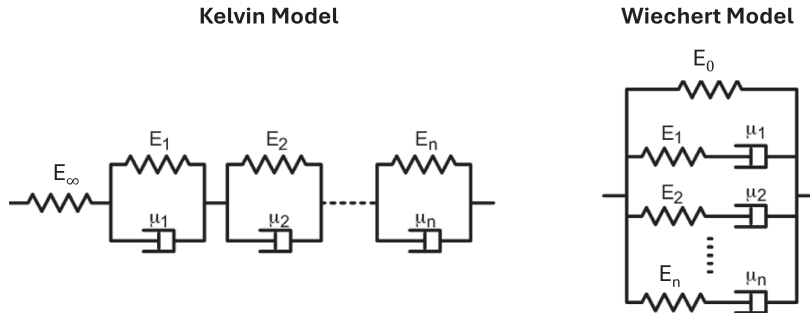


Fig. A.11. The Kelvin model (left) and the Wiechert model (right) for the representation of the mechanical behavior of a viscoelastic material.

A.2. Compliance function in time and frequency domain

Let us now consider to model a viscoelastic material with an infinite series of Voigt elements, in series with a Hookean spring giving the material stiffness for short time, the so-called Kelvin model (see Fig. A.11). This coincides with assuming a continuous distribution of retardation times $L(\tau)$, which is the so-called material retardation spectrum. A general relation for the deformation $\varepsilon(t)$ at time t is given by (Eq. (2.42) in Williams (1964)):

$$\varepsilon(t) = \left\{ C_\infty + \int_0^\infty \frac{L(\tau)}{\left[\frac{d}{dt} + 1/\tau\right] \tau^2} d\tau \right\} \sigma(t), \tag{A.11}$$

where $C_\infty = 1/E_\infty$ is the creep compliance in the glassy limit. Converting Eq. (A.11) in the frequency domain gives:

$$\varepsilon(\omega) = \left\{ C_\infty + \int_0^\infty \frac{L(\tau)}{[i\omega + 1/\tau] \tau^2} d\tau \right\} \sigma(\omega) = \bar{C}(\omega) \sigma(\omega), \tag{A.12}$$

where, i is the imaginary unit and, by definition, $\bar{C}(\omega) = C'(\omega) - iC''(\omega)$ is the complex compliance. Hence, we have:

$$\bar{C}(\omega) = C_\infty + \int_0^\infty \frac{L(\tau)}{[i\omega + 1/\tau] \tau^2} d\tau. \tag{A.13}$$

We note that to match the experimental data, a specific form for the retardation spectrum $L(\tau)$ could be considered. Following Williams (1964) suggestion, a broad-band approximation of the viscoelastic material response can be achieved by using a modified power law for the retardation spectrum, such as:

$$L(\tau) = \left(\frac{C_0 - C_\infty}{\Gamma(n)} \right) \left(\frac{\tau}{\tau_0} \right)^n \exp\left(-\frac{\tau}{\tau_0}\right), \tag{A.14}$$

The complex compliance is defined as follows:

$$\bar{C}(\omega) = C'(\omega) - iC''(\omega), \tag{A.15}$$

where

$$\bar{C}(\omega) = C_\infty + \int_0^\infty \frac{L(\tau)}{[i\omega + 1/\tau] \tau^2} d\tau, \tag{A.16}$$

$$C'(\omega) = C_\infty + \int_0^\infty \frac{L(\tau)}{[1 + \omega^2 \tau^2] \tau} d\tau, \tag{A.17}$$

$$C''(\omega) = \int_0^\infty \frac{L(\tau) \omega}{[1 + \omega^2 \tau^2]} d\tau. \tag{A.18}$$

By using Eq. (A.14) for the retardation spectrum $L(\tau)$ one obtains

$$\bar{C}(\omega) = C_\infty + \frac{(C_0 - C_\infty)}{i\omega\tau_0} \exp\left(-\frac{i}{\omega\tau_0}\right) \mathbf{E}_n\left(-\frac{i}{\omega\tau_0}\right) \tag{A.19}$$

$$C'(\omega) = C_\infty + \frac{(C_0 - C_\infty) (\tau_0\omega)^{-2-n}}{\Gamma(n)} \left\{ \begin{aligned} &\pi (\tau_0\omega)^2 \cos\left(n\frac{\pi}{2} + \frac{1}{\tau_0\omega}\right) \csc(n\pi) + \dots \\ &\dots + (\tau_0\omega)^n \Gamma(-2+n)_p \text{F}_q \left[1; \left\{ \frac{3-n}{2}, 2 - \frac{n}{2} \right\}; -\frac{1}{4(\tau_0\omega)^2} \right] \end{aligned} \right\} \tag{A.20}$$

$$C''(\omega) = \frac{(C_0 - C_\infty) (\tau_0\omega)^{-1-n}}{\Gamma(n)} \left\{ \begin{aligned} &\pi \tau_0 \omega \sin\left(n\frac{\pi}{2} + \frac{1}{\tau_0\omega}\right) \csc(n\pi) + \dots \\ &\dots + (\tau_0\omega)^n \Gamma(-1+n)_p \text{F}_q \left[1; \left\{ 1 - \frac{n}{2}, \frac{3-n}{2} \right\}; -\frac{1}{4(\tau_0\omega)^2} \right] \end{aligned} \right\}, \tag{A.21}$$

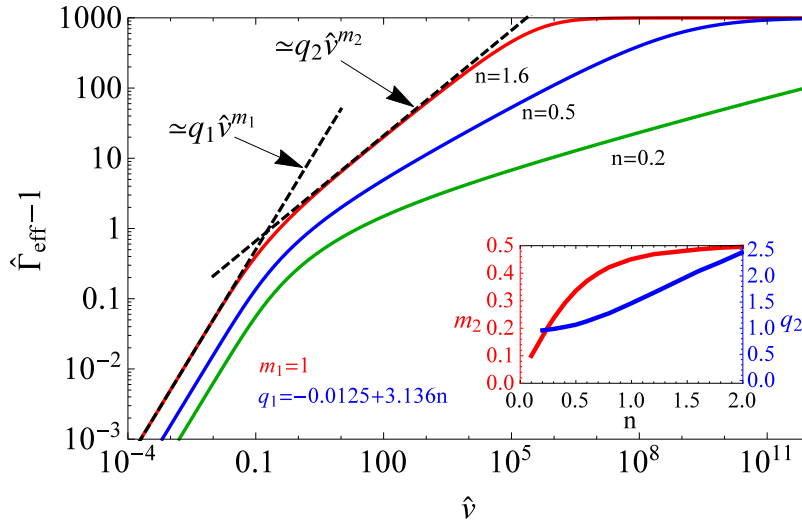


Fig. B.12. Fit of the enhancement of the effective surface energy $\hat{\Gamma}_{eff} - 1$ obtained using a MPL material model in PB theory. Two power law scaling have been identified for $\hat{v} < 1$ and for $\hat{v} > 1$, which coefficients are given in the figure and in the inset as a function of the material exponent n .

where τ_0 is the characteristic time, $n > 0$ is a characteristic exponent, ω is the angular frequency, $C_0 = 1/E_0$ is the relaxed compliance, $C_\infty = 1/E_\infty$ is the instantaneous compliance, ${}_pF_q[a; b; z]$ is the generalized hypergeometric function, $\Gamma(x)$ is the Euler gamma function.

Appendix B. Details of the PB model for the effective surface energy

According to PB theory the dimensionless effective surface energy for a MPL viscoelastic material model $\hat{\Gamma}_{eff}$ is obtained as

$$\hat{\Gamma}_{eff} = \left[1 - (1 - k) I(n, \hat{v}, \hat{\Gamma}_{eff}) \right]^{-1}, \tag{B.1}$$

where $I(n, \hat{v}, \hat{\Gamma}_{eff})$ stands for the integral in Eq. (22), which can be evaluated in closed form as:

$$\begin{aligned} I(n, \hat{v}, \hat{\Gamma}_{eff}) = & \frac{2^{(-3-2n)} \pi^{(-3/2-n)}}{(n-1)\hat{v}} \left\{ -4^{(1+n)} \pi^{(1/2+n)} \left[\hat{\Gamma}_{eff} + \right. \right. \\ & - 2(n-1)\pi \hat{v} {}_pF_q \left[-\frac{1}{2}; \left\{ \frac{1}{2}, \frac{n}{2}, 1 - \frac{n}{2} \right\}; -\left(\frac{\hat{\Gamma}_{eff}}{4\pi\hat{v}} \right)^2 \right] \right] + \\ & + 2\pi \left(\frac{\hat{\Gamma}_{eff}}{\hat{v}} \right)^n \left[4\pi\hat{v}\Gamma \left[1 - \frac{n}{2} \right] {}_pF_q \Big|_{\text{Reg}} \left[\frac{n-1}{2}; \left\{ \frac{1}{2}, \frac{2+n}{2} \right\}; -\left(\frac{\hat{\Gamma}_{eff}}{4\pi\hat{v}} \right)^2 \right] + \right. \\ & \left. + \hat{\Gamma}_{eff}\Gamma \left[\frac{3}{2} - \frac{n}{2} \right] {}_pF_q \Big|_{\text{Reg}} \left[\frac{n}{2}; \left\{ \frac{3}{2}, \frac{(3+n)}{2} \right\}; -\left(\frac{\hat{\Gamma}_{eff}}{4\pi\hat{v}} \right)^2 \right] \right] \right\}, \end{aligned} \tag{B.2}$$

where ${}_pF_q[a, b, z]$ is the generalized hypergeometric function, ${}_pF_q \Big|_{\text{Reg}}[a, b, z]$ is the regularized generalized hypergeometric function and $\Gamma[x]$ is the gamma function (we used Wolfram Mathematica[®] for algebraic manipulation).

To ease the use of Eq. (23) we report here in Fig. B.12 the quantity $\hat{\Gamma}_{eff} - 1$, showing that there exist two power law regimes, the first is a linear scaling where $(\hat{\Gamma}_{eff} - 1) \approx q_1 V^1$, with the coefficient $q_1 = -0.0125 + 3.136n$ depending on the material exponent “ n ”, the second instead can be written as $(\hat{\Gamma}_{eff} - 1) \approx q_2 V^{m_2}$, with the $\{q_2, m_2\}$ constants depending on n , which is shown in the inset of Fig. B.12. Notice that, a SLS would have $n \approx 1.6$ which provides a scaling of $(\hat{\Gamma}_{eff} - 1) \propto V^{0.5}$, while broad band materials provide a much lower exponent m_2 as one can see in Fig. B.12 (inset).

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