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ON THE AFFERRANTE-CARBONE THEORY OF ULTRATOUGH TAPE PEELING

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Abstract. *In a simple and interesting theory of ultratough peeling of an elastic tape from a viscoelastic substrate, Afferrante and Carbone find that there are conditions for which the load for steady state peeling could be arbitrarily large in steady state peeling, at low angles of peeling - what they call "ultratough" peeling (Afferrante, L., Carbone, G., 2016, The ultratough peeling of elastic tapes from viscoelastic substrates, Journal of the Mechanics and Physics of Solids, 96, pp.223-234). Surprisingly, this seems to lead to toughness enhancement higher than the limit value observed in a very large crack in an infinite viscoelastic body, possibly even considering a limit on the stress transmitted. The Afferrante-Carbone theory seems to be a quite approximate, qualitative theory and many aspects and features of this "ultratough" peeling (e.g. conformity with the Rivlin result at low peel angles) are obtained also through other mechanisms (Begley, M.R., Collino, R.R., Israelachvili, J.N., McMeeking, R.M., 2013, Peeling of a tape with large deformations and frictional sliding, Journal of the Mechanics and Physics of Solids, 61(5), pp. 1265-1279) although not at "critical velocities". Experimental and/or numerical verification would be most useful.*

Key Words: *Soft Materials, Peeling, Adhesion, Viscoelastic Materials*

1. INTRODUCTION

Adhesion of two objects can arise due to chemical bonding as by a glue, or by physical interaction such as by van der Waals or electrostatic attraction [1]. Various tests have been devised to probe the strength and toughness of adhesive bonds, including the

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peeling of a tape from a substrate or from another tapelike feature [2, 3]. Our schematic of the former version of the test is illustrated in Fig. 1. Rivlin [2] provided a formula for the relationship between the effective adhesion energy, Γ , and the force, P , required to peel an inextensible tape from an elastic substrate. The energy of adhesion as the work per unit area of bond requires to progressively separate the objects from each other. Kendall [4, 5] then augmented the formula by accounting for the effect of the strain energy of an elastic tape. The importance of adhesion, and its science and technology, can be confirmed by perusal of the references provided by Afferrante and Carbone [6], and by the reviews by Creton and Ciccotti [7] and Long et al. [8].

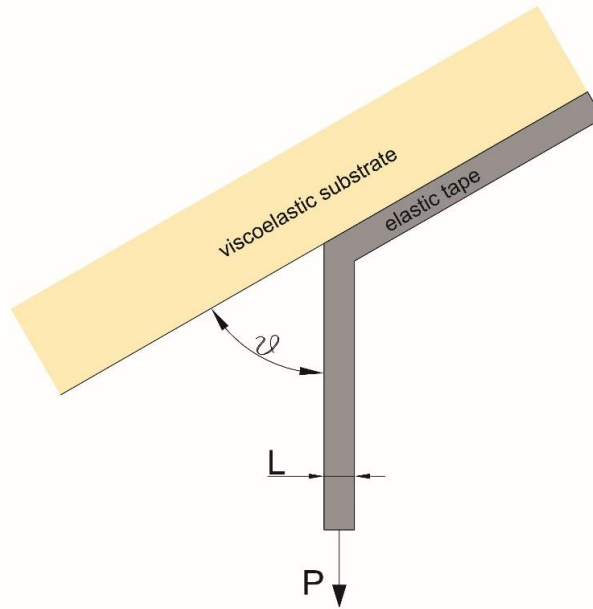


Fig. 1 Geometry of the peeling of an elastic tape from a viscoelastic substrate for the AC theory

Viscoelastic effects are known to occur in the peeling of polymer and organic materials, e.g. Kendall [2, 3] presents such data and attributes the effect to viscoelastic behavior in the separation process. Such viscoelastic behavior is addressed in an extensive literature (e.g. [9–12]), suggesting steady state critical crack growth during peeling occurs with an enhanced apparent work of adhesion according to the Gent-Schultz law

$$\frac{\Gamma}{\Gamma_0} = 1 + \left(\frac{v}{v_0} \right)^n \quad (1)$$

where Γ is the apparent or effective work of adhesion, Γ_0 is its reference or datum value at a very slow rate of propagation, v is the rate of peeling, and $v_0 = (ka_T^n)^{-1}$ in which k and n

($0 < n < 1$) are (or are supposed to be) materials constants, and a_T^n is the WLF factor for unifying results from various temperatures (Williams, Landel and Ferry [13]).

Gent and Schultz [10] describe the reference value, Γ_0 , as the thermodynamic work of fracture, and attribute the enhancement in Eq. (2) to viscoelastic dissipation. A model incorporating such a point of view was developed by de Gennes [14], described as the “viscoelastic trumpet,” with different mechanisms of deformation and adhesive rupture dominating at different scales at different rates of crack or peel front propagation. However, de Gennes [14] finds that the resulting highest effective work of adhesion for very rapid crack or peel front propagation is finite instead of unlimited as in Eq. (1), and is given by

$$\frac{\Gamma_\infty}{\Gamma_0} = \frac{E_\infty}{E_0} \quad (2)$$

where E_∞ is the instantaneous modulus and E_0 is the relaxed, or equilibrium modulus. Note that the ratio in Eq. (2) can span 3 or 4 orders of magnitude [12].

The velocity dependence of Eqs. (1) and (2) was, at first, difficult to rationalize as cracks and peel fronts in linear viscoelastic materials admit an inverse square root stress singularity, implying that the high strain rate modulus should always dominate at the crack and peel front tip, indicating a lack of velocity dependence [15]. However, introduction of a Barenblatt or Maugis-Dugdale cohesive zone at the crack tip enabled the identification of velocity dependence for crack propagation [16-20], as then the effective modulus at the crack tip depends on the propagation rate. The resulting model predicts the relationship in Eq. (2) for Mode I crack propagation (i.e. tension), where, in this case, Γ_∞ is defined to be K_∞^2 / E_0 , where K_∞ is the Mode I stress intensity factor at high rates of crack propagation. Similarly, $\Gamma_0 = K_0^2 / E_0$.

One can expect an interplay among cohesive zone behavior, peel rate, viscoelastic dissipation and specimen sizescale. Such an effect was explored in finite element modeling by Rahul Kumar et al. [21] for T-peeling of a viscoelastic polymer strip. They found that the apparent adhesion energy at low and high rates of peeling effectively obeys Eq. (2), but with an intermediate propagation rate where viscoelastic dissipation additionally enhances the apparent adhesion energy. As a result, the relationship between the apparent adhesion energy and the peel rate is non-monotonic, in contrast to Eq. (1). Afferrante and Carbone [6] (henceforth AC) address a similar issue but for an elastic tape being peeled from a viscoelastic substrate as in Fig. 1. The substrate is considered to be semi-infinite, the tape thickness is L and the peel angle is θ . To model the transfer of loads to the substrate, AC assumed that they could be treated as uniform tractions applied over a width L at the peel front, i.e. a traction normal to the substrate surface plus a shear traction. The substrate surface is traction free elsewhere, allowing use of the viscoelastic correspondence principle for the substrate. The AC model therefore has features that make it similar to a cohesive zone model with uniform cohesive tractions, and has a size scale, the elastic tape thickness, that can introduce an interplay between the peel rate and viscoelastic dissipation in the substrate. In the next section we identify some aspects of the peeling behavior in the AC model.

2. AFFERRANTE-CARBONE MODEL

For an elastic tape of modulus E and a viscoelastic substrate with a single relaxation time τ_0 , (e.g. the standard model of linear viscoelasticity), the AC equation (14) [6] provides

$$\frac{\Gamma_0}{EL} = \left[\frac{1}{2} - \frac{E}{E_0} f_v \left(\frac{v\tau_0}{L} \right) \right] \left(\frac{P}{EL} \right)^2 + \left(\frac{P}{EL} \right) (1 - \cos \vartheta) \quad (3)$$

where P is the load per unit out-of-plane width of the tape and f_v is a function of exponential integrals of its argument $v\tau_0/L$, see the AC equation (15) [6]. Note that Eq. (3) is written as an expression for the intrinsic adhesion energy, Γ_0 . The function f_v is zero for both very slow peeling rate, $v \rightarrow 0$, and at extreme rates of peeling, $v \rightarrow \infty$. Otherwise, f_v is positive, i.e. consistent with viscoelastic dissipation from which it arises, and has a maximum at an intermediate rate of peeling that depends weakly on E_∞/E_0 if $E_\infty/E_0 > 15$. At very low peel rates, and very high peel rates, with $f_v \rightarrow 0$, the result in Eq. (3) is simply Kendall's [4], or Rivlin's [2] if the strain energy term containing P^2 is neglected, as it can be for high peel angles. This behavior is expected as the system behaves purely elastically at very low and very high rates of peeling with negligible viscoelastic dissipation. As a result, the formula in Eq. (3) at very slow and very fast rates of peeling shows that in these cases the intrinsic adhesion energy is the only contributor to the work of adhesion. However, the intrinsic adhesion energy may be rate dependent, as in the case where a soft, lightly cross-linked polymer is being used as an adhesive and its crazing and fibrillation can lead to viscoelastic behavior within the adhesive.

The result in Eq. (3) is a quadratic equation for P/EL with discriminant

$$\Delta = (1 - \cos \vartheta)^2 + \frac{4\Gamma_0}{EL} \left[\frac{1}{2} - \frac{E}{E_0} f_v \left(\frac{v\tau_0}{L} \right) \right] \quad (4)$$

and, therefore, real solutions for P/EL exist only for $\Delta \geq 0$. The solutions are

$$\frac{P}{EL} = \frac{1 - \cos \vartheta \pm \sqrt{\Delta}}{1 - \frac{2E}{E_0} f_v \left(\frac{v\tau_0}{L} \right)} \quad (5)$$

and we accept only positive real solutions. We deduce that the apparent adhesion energy, Γ , the sum of the thermodynamic adhesion energy and the contribution of viscoelastic dissipation, is given by

$$\frac{\Gamma}{EL} = \frac{1}{2} \left(\frac{P}{EL} \right)^2 + \left(\frac{P}{EL} \right) (1 - \cos \vartheta) = \frac{\Gamma_0}{EL} + \frac{E}{E_0} f_v \left(\frac{v\tau_0}{L} \right) \left(\frac{P}{EL} \right)^2 \quad (6)$$

Note that the first result on the right-hand side of Eq. (6) is the normalized energy release rate as given by Kendall [4]. We observe, therefore, that, according to the AC model, the Kendall [4] formula for the energy release rate also provides the effective adhesion energy, as expressed by the first equality of Eq. (6).

From Eq. (6) we obtain the ratio

$$\frac{\Gamma}{\Gamma_0} = 1 + \frac{EL}{\Gamma_0} \frac{E}{E_0} f_v \left(\frac{v\tau_0}{L} \right) \left(\frac{P}{EL} \right)^2 \quad (7)$$

where P is obtained from Eq. (5) when there are real, positive solutions for it.

For illustration, we consider the case where $E/E_0=1.5$ and $E_\infty/E_0=10$, $\Gamma_0/EL=10^{-4}$ as also used by AC. In Fig. 2a, we present the result for Γ/Γ_0 from Eq. (7) where the lower positive real result for P/EL from Eq. (5) is utilized. It can be seen that the result for $\theta=\pi/64=2.8^\circ$ can have an enhancement of the apparent adhesion energy that can be very large. Although not shown in Fig. 2a, the result for Γ/Γ_0-1 for $\theta=\pi/64=2.8^\circ$ can be as high as ≈ 140 . There are no real solutions for this angle in the range where the results in black are not visible in the plot. This outcome is much larger than $E_\infty/E_0=10$ as predicted by de Gennes [14] for the ratio of the apparent adhesion energy for very fast peeling compared to very slow peeling. The enhancement of the apparent adhesion energy is thus non-monotonic and has a maximum at peeling rates for which $v\tau_0/L \approx 1$.

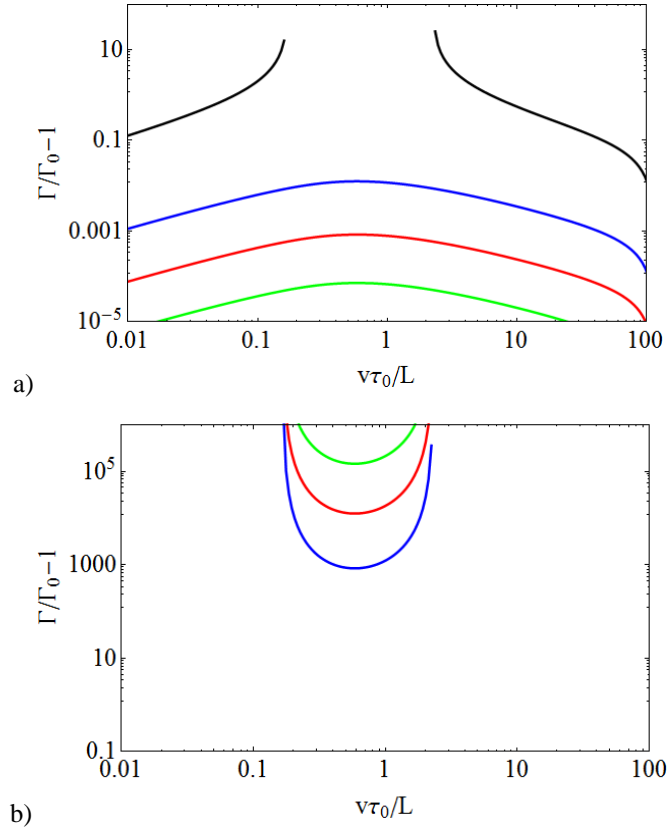


Fig. 2 Enhancement (Γ/Γ_0-1) for the AC peeling theory shown as a function of the peeling velocity v : (a) lower load solution (b) higher load solution, both for $E/E_0=1.5$, $\Gamma_0/EL=10^{-4}$, $E_\infty/E_0=10$. Peel angles are $\theta = \pi/64, \pi/8, \pi/4, \pi/2$ (respectively black, blue, red and green).

Note also that for peel angles equal to $\vartheta = \pi/8 = 22.5^\circ$ and above the enhancement of the apparent adhesion energy is negligible. We note that we find negligible enhancement of the apparent adhesion energy for peel angles greater than approximately 8° for the parameters used to plot Fig. 2a. As noted above, the effective adhesion energy is then essentially equal to the intrinsic adhesion energy, Γ_0 , and is so over the whole range of peel rates. As also noted above, Γ_0 itself may be rate dependent, leading to a higher peel force at fast rates of peeling than at low rates. However, it is then not obvious that Eq. (2) will predict the ratio of effective adhesion energies at fast peel rates versus slow rates, and the behavior in terms of effective adhesion energy may well be given by Eq. (1). If the intrinsic adhesion energy lacks rate dependence, then the peel force for peel angles equal to or greater than 22.5° will be insensitive to the rate of peeling, in disagreement with Eqs. (1) and (2). For example, if van der Waals forces are in control of adhesion, according to the AC model, the peel force can be expected to be insensitive to the rate of peeling for peel angles greater than or equal to 22.5° .

As noted by AC, it is possible that the higher magnitude, 2nd solution of Eq. (5) is relevant during peeling and that the system can be forced into this regime. To explore this possibility, in Fig. 2b we use the higher magnitude, 2nd solution from Eq. (5) to evaluate and plot the result from Eq. (7) for the same parameters used for Fig. 2a. We plot only some of the lower values we obtain as many of the values are extremely high for the parameters utilized. Given that, for peel angles greater than or equal to 22.5° the 2nd, higher magnitude solution gives an apparent adhesion energy that is at least 3 orders of magnitude greater than the lower magnitude solution, we conclude that it is unlikely that, for the parameters used in Fig. 2, that the 2nd, higher magnitude solution is relevant to peeling. There are no real solutions in Fig. 2b for the $\vartheta = \pi/64$ case.

We also find that the high apparent adhesion energy regime of peeling in the AC model depends on the ratio E/E_0 . As noted above, when $E/E_0 = 1.5$, such high apparent adhesion energy behavior occurs for peel angles below 8° . For $E/E_0 > 1.5$ this range of ultratough adhesion becomes larger.

3. DISCUSSION

Compared to other widely used models, like the Gent-Schultz law (1), and the ‘viscoelastic trumpet’ developed by de Gennes, the AC point of view appears to be, like them, an approximate solution. Indeed, the first two give results that are essentially qualitative, and whose validity depends on experimental databases; this point is clearly stated, e.g., by Saulnier et al. [22] in the case of the viscoelastic trumpet. On the other hand, the AC model is developed following the mathematical consequences of an energy balance imposed through a well-defined, though inexact, physical model. Notwithstanding the great difference between the respective frameworks, the AC results appear to be qualitatively similar to the outcomes of Saulnier et al. [22]. In fact, in [22] the energy balance is based on a singular stress field directly derived from the correspondence principle, where such a singular stress is completely absent in AC. More specifically, both models predict that the viscous contribution to the detachment energy rate, $\Gamma(\dot{\nu})$, is zero for very low or very high peel rates, whereas it can be very large at an intermediate rate. At this intermediate rate the AC results seem to be in contrast with the experimental

outcomes from peel experiments in geometries similar to Fig. 1 with viscoelastic components.

It seems quite likely that the AC model is oversimplified as the interaction between the tape and the substrate is modeled as a uniform traction within a constant interaction length L , whatever the traction angle ϑ , the tape and substrate stiffness and the tape thickness. To explore this point, let us consider two extreme conditions $\vartheta = \pi/2$ and $\vartheta = 0$ (see Fig. 3). In the first case, the AC model neglects the tape flexural stiffness (although if the tape is very thin its flexural stiffness will be negligible so this is not always a problem), and in the second one, there is an issue about the interaction zone length: the cohesive zone length could be much smaller than L and it might be larger than L . Also, if it is friction that determines the interaction length frictional slip zone might be larger than L and it might be smaller than L depending on the friction shear stress. If it is sticking friction, the interaction zone will be very small. On the other hand, Eq. (7) shows clearly that the interaction length L is a very important parameter in the evaluation of the viscous contribution to the apparent adhesion energy.

For these reasons, one should consider the AC predictions to be only approximate and qualitative, especially for low peel angles. Indeed, it seems very unlikely that ‘ultratough peeling’, even if it exists, can be captured with such simplistic assumptions as are utilized by AC.

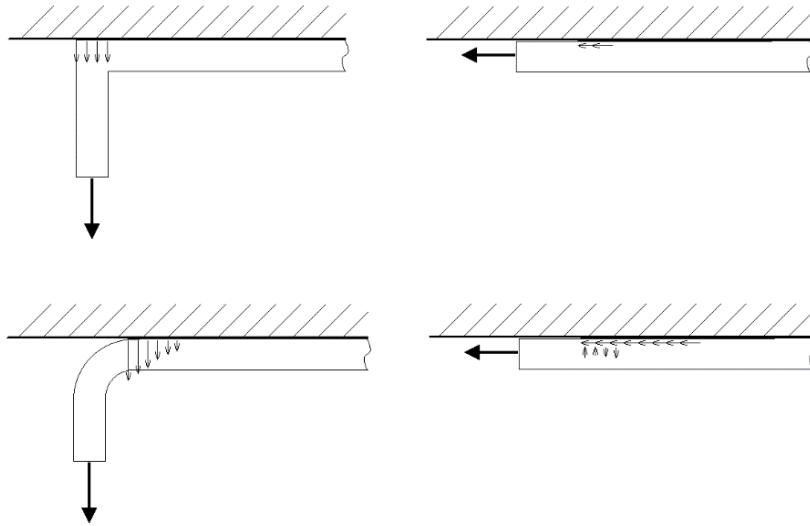


Fig. 3 Interface interaction for the AC model for $\vartheta = \pi/2$ and $\vartheta = 0$ (above) with a more realistic one shown below

Even if the AC result is a straightforward consequence of the energy balance (and a poorly designed model), the interpretation of Eq. (7) as a ‘toughness’ may be misleading, notwithstanding its coherence with many widely accepted formulations. This implies that, in some conditions, the power absorbed by the viscous substrate in steady state peeling is unlimited, which is physically an unlikely result. Instead of identifying toughness, as is normally what is done using Kendall [5], in the AC case one can speak unambiguously of

peel load and, ultimately, of ‘ultra-high-strength’ peeling. While AC provides a result that we find to be counter-intuitive, unfortunately we cannot yet present a better model. Thus, we can neither prove our claims nor disprove AC.

There is a substantial difference between the AC and other models cited. This situation may partially be attributed to the fact that the volume in which viscous energy is dissipated in the AC model is infinite. A more detailed model, consistent with physical behavior of the interface interaction between the tape and the substrate would help deeper investigation.

4. CONCLUSIONS

We have discussed the fact that the "ultratough peeling" theory of Afferrante and Carbone [6] leads to two possible equilibrium solutions for the load at a given peeling speed. The low load solution in some regimes produces very high toughness enhancement, possibly already much larger even than that expected in a crack in an infinite system. More problematic appears to be the high load solution which, even considering a limit stress carried by tape and substrate, will not avoid some solutions to have unbounded toughness enhancement. In addition, viscoelastic effects seem to induce an on-off mechanism, where for large angles of peeling it predicts solutions extremely close to the purely elastic Kendall case, while below a threshold angle, the toughening abruptly rises to very high levels, and the "ultratough" peeling regime appears. It seems very likely that this strange result is due to approximations present within the model, even if it is quite sophisticated from a mathematical point of view. Features of this "ultratough" peeling (e.g. conformity with the Rivlin result at low peel angles) are obtained also through other mechanisms [23] although not at “critical velocities”.

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