

Energetic criterion for adhesion in viscoelastic contacts with non-entropic surface interaction

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ABSTRACT

We suggest a detachment criterion for a viscoelastic elastomer contact based on Griffith's idea about the energy balance at an infinitesimal advancement of the boundary of an adhesive crack. At the moment of detachment of a surface element at the boundary of an adhesive contact, there is some quick (instant) relaxation of stored elastic energy which can be expressed in terms of the creep function of the material. We argue that it is only this "instant part" of stored energy which is available for doing work of adhesion and thus it is only this part of energy relaxation that must be used in Griffith's energy balance. The described idea has several restrictions. Firstly, in this pure form, it is only valid for adhesive forces having an infinitely small range of action (which we call the JKR-limit). Secondly, it is only applicable to non-entropic (energetic) interfaces, which detach "at once" and do not possess their own kinetics of detachment.

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1. Introduction

Adhesion of elastic bodies can be understood and described using the principle of virtual work. For non-dissipative systems it can be used in the form of "energy balance", first suggested by Griffith (1921) and used in the theory of Johnson, Kendall and Roberts (1971), (Popova & Popov, 2018). This approach cannot be applied to situations where dissipative forces are in play, in particular to tangential adhesive contacts and to adhesive contacts of viscoelastic bodies. However, the principle of energy balance is so simple and attractive that it is used even if is not applicable, by introducing an effective work of adhesion and postulating its dependence on the crack propagation velocity (see e.g. Barquins & Maugis, 1981). We would like to revisit this approach and to argue that it might be possible to apply the energy balance principle in a pure and exact way to viscoelastic adhesive contacts. If the characteristic time of stress relaxation in the elastomer is much larger than the characteristic time of detachment, then one could think of detachment as a practically instant process. If the loaded surface of a viscoelastic body is instantly unloaded, then there is always some instant elastic relaxation followed by slow relaxation depending on the detailed rheology of the medium. The detachment can only occur if the instant relaxation part of the elastic energy is equal to the work of separation of surfaces. This means that the energy balance can be used for viscoelastic bodies too, in a modified form.

It is important to clearly understand that in viscoelastic contacts there can be two independent kinetic processes and correspondingly two independent "rheologies": the first one related to the internal processes in the volume of the material and the second one related to the process of detachment. For example, if two

elastomers are in contact, one can imagine that their molecules are tangled up and must be disentangled to separate. This is a process that might take some time and might be strongly dependent on the temperature. On the other hand, if an elastomer is in contact with a smooth solid, this disentanglement does not occur, and the separation occurs just by overcoming some critical stress. One can say that the first type of interfaces is viscoelastic and the second one elastic (even while the volume properties remain viscoelastic in both cases). In analogy with material classes, we can speak of "energetic" (elastic) and "entropic" (viscoelastic) surface interactions (Figure 1). Under energetic interactions we understand interactions dominated by the potential interaction energy.

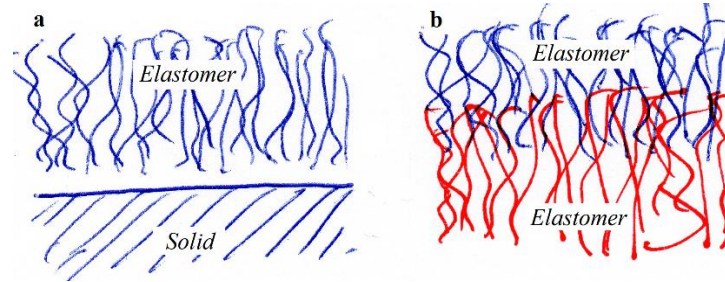


Figure 1. Energetic (a) and entropic (b) surface interactions

In the present paper only the case of energetic interfaces will be considered, as it allows to more clearly describe the suggested idea. The detachment criterion will be explained in the following using the Method of Dimensionality Reduction (MDR). Normally, one discusses the crack propagation in terms of the stress concentration factors. But it can be equivalently described in the framework of the Method of Dimensionality Reduction. While the results obtained in the direct three-dimensional presentation and the MDR are equivalent, the great difference is in the simplicity of thinking and analysis. The MDR-presentation is especially useful in the case where there is a scale separation of processes in the considered system. In the real space presentation, there is a singularity at the edge of the crack, so the scale separation is not "visible" (as all processes are infinitely rapid). In the MDR, on the contrary, no singularity exists, and the scale separation becomes obvious. In Section 2, we briefly describe the main calculation steps of the MDR.

2. Solution of the adhesive normal contact problem of elastic bodies in the framework of the Method of Dimensionality Reduction

The calculation method via the MDR consists of the following steps (Popov, 2017): In the first step the given three-dimensional profile $\tilde{z} = f(r)$ is transformed into an equivalent plane profile $g(x)$ via equation:

$$g(x) = |x| \int_0^{|x|} \frac{f'(r)}{\sqrt{x^2 - r^2}} dr \quad (1)$$

(going back to the theory by Schubert (Popova & Popov, 2020)). The profile $g(x)$ is now pushed into the one-dimensional elastic foundation, a series of springs with the spacing Δx and stiffness

$$\Delta k_z = E^* \Delta x, \quad (2)$$

until a contact radius a is reached, where the effective elasticity modulus E^* is defined as:

$$\frac{1}{E^*} = \frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2}. \quad (3)$$

This process is depicted in Figure 2 on the left. In the third step, the indenter is lifted up. It is assumed that all springs involved in the contact adhere to the indenter - the contact radius thus remains constant. In this process, the springs at the edge experience the maximum increase in tension. Upon reaching the maximum possible elongation of the outer springs

$$\Delta l(a) = \sqrt{\frac{2\pi a \Delta \gamma}{E^*}} \quad (4)$$

they detach. This criterion was discovered by Heß (2011) and is known as the *rule of Hess*.

This rule can be easily derived using the principle of virtual work. According to this principle, the system is in equilibrium when the energy does not change for small variations of its generalized coordinates. Applied to the adhesive contact, it means that the change in the elastic energy for a small reduction of the contact radius from a to $a - \Delta x$ is equal to the change in the surface energy $2\pi a \Delta x \Delta \gamma$, where $\Delta \gamma$ is the separation work of the contacting surfaces per unit area. Since the MDR maps the relation of force to displacement exactly, the elastic energy is also reproduced exactly. The change in the elastic energy can, therefore, be calculated directly in the MDR model. Due to the detachment of each one spring at the edge of the contact, the elastic energy is reduced by $E^* \Delta x \Delta l^2$. Balance of the changes in the elastic and the adhesive energy results in

$$2\pi a \Delta x \Delta \gamma = E^* \Delta x \Delta l^2 \quad (5)$$

which results in (4).

The corresponding equilibrium described by the three quantities (F, d, a) provides the exact solution to the adhesive contact problem. The displacement of the outer springs is negative, with the absolute value equal to the critical value: $w_{1D}(a) = -\Delta l(a)$. It follows that

$$d = g(a) - \Delta l(a). \quad (6)$$

The normal force is given by the equation

$$F_N = 2E^* \int_0^a [d - g(x)] dx. \quad (7)$$

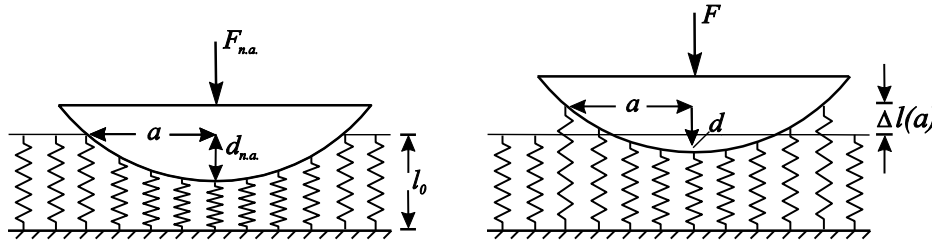


Figure 2. Representation (in the framework of MDR) of the indentation and lifting process of a spherical 1D-indenter with an elastic foundation, which exactly models the properties of the adhesive contact between a rigid spherical indenter and an elastic half-space.

Note that the same energetic balance method was used in (Pohrt & Popov, 2015) and (Popov et al., 2017) for deriving the detachment criterion of single simulation cells in the Boundary Element Method.

3. Energetic detachment criterion in the case of a viscoelastic material

The Method of Dimensionality Reduction can be applied also to viscoelastic contacts by replacing "springs" with corresponding rheological elements as defined in (Popov et al., 2018). This representation has the form:

$$\begin{aligned} \Delta k_z(t) &= \frac{2}{1-\nu} G(t) \Delta x = 4G(t) \Delta x, \\ \Delta k_x(t) &= \frac{4}{2-\nu} G(t) \Delta x = \frac{8}{3} G(t) \Delta x, \end{aligned} \quad (8)$$

in the time domain, where $G(t)$ is the time dependent shear modulus, or in the frequency domain,

$$\begin{aligned} \Delta \hat{k}_z(\omega) &= 4\hat{G}(\omega) \Delta x, \\ \Delta \hat{k}_x(\omega) &= \frac{8}{3} \hat{G}(\omega) \Delta x \end{aligned} \quad (9)$$

with $\hat{G}(\omega)$ being the complex frequency dependent shear modulus.

Linear rheology can always be represented by a generalized Maxwell element consisting of a soft spring and a series of Maxwell elements with different stiffness and damping connected in parallel (relaxation element, Figure 3a) or as a series of Kelvin elements connected in series (creep element, Figure 3b). In the general discussion, we will use the second presentation.

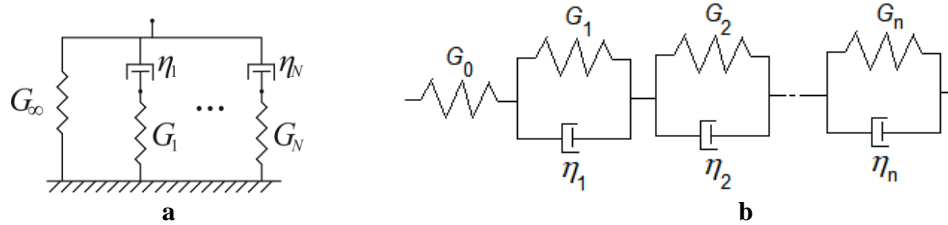


Figure 3. Generalized Maxwell element (a) and generalized relaxation element (b) for simulation of an arbitrary linear rheology. The elements are equivalent if the parameters are chosen correspondingly.

For viscoelastic rheology, each "spring" in Figure 2 is replaced by a creep element shown in Figure 3b. If the body is lifted, the element at the edge of the contact will have an elongation Δl . However, now this elongation is a sum of elongations of all "subelements" of the creep element, $\Delta l = \Delta l_0 + \Delta l_1 + \dots + \Delta l_n + \dots$, where Δl_0 is the elongation of the spring G_0 , Δl_1 the elongation of the Kelvin-Element (G_1, η_1) , and so on. Now let us assume that the element detaches from the indenter. Some part of the energy will relax very quickly, "at once". This is the part which is stored in the spring G_0 . And only this part can be "used" for the formation of new surfaces, for the work of adhesion. The elastic energy stored in all Kelvin elements will need some time to be relaxed. But the detachment occurs at a molecular scale, and thus, from a macroscopic point of view, instantly. The detachment can occur only if the elastic energy which is available "at once" is enough for doing the work of adhesion. Further, as the instant reaction of a viscoelastic medium is purely elastic, we are therefore empowered to use the energy balance and to equate the energy $2 \cdot (4G_0) \Delta x \frac{\Delta l_0^2}{2}$ to the work of adhesion, $2\pi a \cdot \Delta x \cdot \Delta \gamma$, where $\Delta \gamma$ is the specific work of adhesion:

$$2\pi a \cdot \Delta x \cdot \Delta \gamma = (4G_0) \Delta x \Delta l_0^2, \quad (10)$$

which looks exactly like the detachment criterion (5) for the elastic case, with the only difference that G_0 is now the *glass modulus* of the medium and Δl_0 is only part of the total elongation. Note that we consider here the case of energetic interfaces, thus we assume that $\Delta \gamma$ is a well-defined material parameter which does not depend on the velocity of detachment. How large the elongation part Δl_0 is depends on the loading history. Solving Eq. (10) with respect to Δl_0 gives

$$\Delta l_0 = \sqrt{\frac{\pi a \Delta \gamma}{2G_0}}. \quad (11)$$

4. Detachment criterion for viscoelastic media in the Boundary Element Method (BEM)

Let us consider the simplest discretization of the contacting surfaces consisting of square elements with the side length Δ as shown in Figure 4. The complete procedure of BEM for the non-adhesive contact of viscoelastic materials is described in (Kusche 2016). In each calculation iteration of the BEM, the stress and displacement of each particular discretization element are determined, and it is decided if the element should still remain in contact. For non-adhesive contacts this is the case if the pressure remains positive. In an adhesive contact, pressures may become negative and thus a more elaborate rule of detachment is needed. Pohrt & Popov (2015) suggested to make the decision about detachment of a single element based on the Griffith' energy criterion: the element would detach if the energy released by its detachment exceeds the work of adhesion. As

the instant reaction of the medium to the sudden vanishing of stress is purely elastic, we can calculate the instant relaxation energy using the theory of elasticity (Pohrt & Popov, 2015):

$$U_{el}(\tau) = \frac{\sigma^2}{2\pi E^*} \int_0^{\Delta} \int_0^{\Delta} \int_0^{\Delta} \int_0^{\Delta} \frac{1}{\sqrt{(x-\tilde{x})^2 + (y-\tilde{y})^2}} d\tilde{x}d\tilde{y}dx dy = \frac{\sigma^2}{E^*} \chi, \quad (12)$$

where

$$\chi = \Delta^3 \frac{2}{3\pi} \left(1 - \sqrt{2} + \frac{3}{2} \log \left(\frac{\sqrt{2}+1}{\sqrt{2}-1} \right) \right) \approx 0.473201 \Delta^3. \quad (13)$$

Equating the elastic energy to the work of adhesion,

$$U_{el} = U_{adh} = (\Delta\gamma) \Delta^2, \quad (14)$$

we come to the following criterion for detachment:

$$\sigma_c = -\sqrt{\frac{E^* \Delta\gamma}{0.473201 \cdot \Delta}}. \quad (15)$$

In the above equations, we use the elastic modulus E^* responsible for the instant relaxation, which is the glass modulus. For an incompressible medium $E^* = 4G_0$ and we finally find

$$\sigma_c = -\sqrt{\frac{4G_0 \Delta\gamma}{0.473201 \cdot \Delta}}. \quad (16)$$

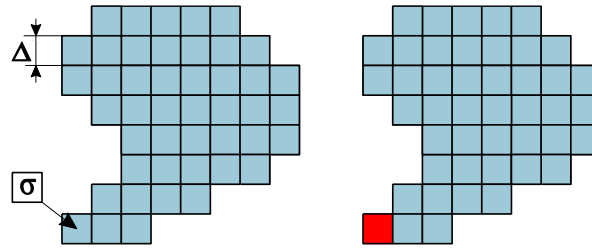


Figure 4. In each calculation step, stress in each particular discretization element is determined. If the stress σ in a given element at the boundary of the contact area exceeds the critical value, it is “detached” and the stress in this element is set to zero.

Thus, the detachment criterion *coincides* with that found by Pohrt & Popov (2015), with the only difference that the modulus which should be used is the *glass* modulus.

5. Detachment of a parabolic indenter from a viscoelastic half-space described by the "Standard model"

We now illustrate the idea described above on an example of an adhesive contact between a rigid parabolic indenter and a viscoelastic medium with rheology described by the "standard model", illustrated in Figure 5.

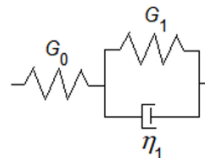


Figure 5. "Standard model" for an elastomer.

Consider a rigid parabolic indenter having the shape $z = r^2 / (2R)$, where r is the in-plane polar radius and R the radius of curvature. The indenter is first pressed into the viscoelastic medium to the depth d_0 and remains in that state for a long enough time, so that all relaxation processes can be considered as completed.

After that, the indenter is lifted with a constant velocity v_0 . To solve this problem, we again use the method of dimensionality reduction. According to (1), the three-dimensional profile $r^2 / (2R)$ is replaced by the plane profile $g(x) = x^2 / R$. For all points of the viscoelastic foundation which are in contact with the indenter, the vertical displacement is given by the equation

$$u_{1D,z}(x) = d(t) - x^2 / R = d_0 - v_0 t - x^2 / R. \quad (17)$$

This displacement is the sum of the elongation u_{G_0} of the spring G_0 and u_{G_1} of the spring G_1 :

$$u_{1D,z}(x) = u_{G_0} + u_{G_1}. \quad (18)$$

The force equilibrium at the connection point between the spring G_0 and the Kelvin element (G_1, η_1) reads

$$G_0 u_{G_0} = G_1 u_{G_1} + \eta_1 \dot{u}_{G_1}. \quad (19)$$

From (17), (18) and (19), it follows that

$$d_0 - v_0 t - \frac{x^2}{R} = \left(1 + \frac{G_1}{G_0}\right) u_{G_1} + \frac{\eta_1}{G_0} \dot{u}_{G_1}. \quad (20)$$

The initial condition for this equation follows from (19), (18), (17) by setting $\dot{u}_{G_1} = 0$:

$$d_0 - \frac{x^2}{R} = \left(1 + \frac{G_1}{G_0}\right) u_{G_1} \Big|_{t=0}. \quad (21)$$

The general solution of Eq. (20) with initial condition (21) is

$$u_{G_1} = At + B + Ce^{-t/\tau}, \quad (22)$$

where

$$A = -\frac{v_0}{1 + G_1 / G_0}, \quad B = \frac{1}{1 + G_1 / G_0} \left[d_0 - \frac{x^2}{R} + \tau v_0 \right], \quad \tau = \frac{\eta_1}{G_0 + G_1}, \quad C = -\frac{\tau v_0}{1 + G_1 / G_0}. \quad (23)$$

Correspondingly,

$$u_{G_0} = d_0 - v_0 t - \frac{x^2}{R} - At - B - Ce^{-t/\tau} = \frac{1}{G_0 + G_1} \left[G_1 \left(d_0 - v_0 t - \frac{x^2}{R} \right) + G_0 \tau v_0 e^{-t/\tau} \right]. \quad (24)$$

The detachment criterion (11) for springs at the edge of the contact, $x = a$, reads

$$-\left[\left(d_0 - v_0 t - \frac{a^2}{R} \right) + \frac{G_0}{G_1} \tau v_0 e^{-t/\tau} \right] = \frac{G_0 + G_1}{G_1} \sqrt{\frac{\pi a \Delta \gamma}{2 G_0}}. \quad (25)$$

This equation gives an implicit dependency of the contact radius on time.

Quasistatic limiting case

In the limiting case of very small pulling velocity, the term with the exponential function can be neglected, and the detachment criterion takes the form

$$\frac{a^2}{R} - (d_0 - v_0 t) = \frac{G_0 + G_1}{G_1} \sqrt{\frac{\pi a \Delta \gamma}{2 G_0}}. \quad (26)$$

Assuming $G_0 \gg G_1$, this equation can be rewritten as

$$\frac{a^2}{R} - d(t) = \sqrt{\frac{\pi a (\Delta \gamma G_0 / G_1)}{2 G_1}}. \quad (27)$$

This is the standard JKR equation for a medium with the effective elastic modulus $E^* = 4G_1$ and effective work of adhesion $\Delta\gamma_{\text{eff}} = \Delta\gamma G_0 / G_1$, which is much larger than the "true" work of adhesion $\Delta\gamma$.

6. Discussion and conclusion

The intention of the present paper was not to solve a particular problem in the theory of adhesion but to raise a question: Is it possible to apply the energy balance by Griffith to adhesive contacts of viscoelastic materials? We argue that this might be possible, at least for a limited class of interface interactions, namely the "non-entropic" or "energetic" interfaces. The possibility of applying the energy balance to viscoelastic crack propagation is based on the well-known fact, that viscoelastic media when loaded or unloaded, always show a very rapid, pure elastic reaction followed by a longer viscoelastic relaxation. We argue that it is only this instant part of the energy relaxation which has to be equated to the work of adhesion. The argument is based on the consideration of scale separation: The detachment of elastic interfaces occurs (from a macroscopic point of view) instantly, and thus only the energy which can be released at this time scale is relevant. We have illustrated this idea for the case of quasistatic detachment of a viscoelastic solid and have shown, that even in this case the bodies cannot be considered as purely elastic, as the energy released at detachment is only a small part of the energy stored in the system. However, the JKR theory remains valid in this limiting case with the only correction that the true work of adhesion has to be replaced by a (much larger) effective work of adhesion. Formulation of the detachment criterion for this class of surfaces for the Boundary Element Method is also straightforward. In the general case, it seems that the concept of an "effective work of adhesion" is not applicable. We hope that this new view on the mechanics of viscoelastic cracks may contribute to the solution of some controversies in the theory of viscoelastic adhesive contacts (see e.g. Ciavarella et al., 2021).

Finally, we would like to stress that the idea suggested in the present paper applies only to energetic interfaces and the "JKR-limit" (in the sense of infinitely small interaction range of adhesive forces). The cases of finite interaction range of adhesive forces (Derjagin et al. 1975) needs an additional consideration.

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