

# Kinetics of the Spontaneous Peeling of Elastomers

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## Synopsis

The spontaneous delamination of an elastomer strip in contact on a deformable substrate is studied. Theoretically, the system is analysed in terms of the energy balance theory of fracture by the general equation  $G - w = w \cdot \Phi(a_0 \cdot v)$ , proposed by Maugis and Barquins in 1978, in which  $G$  is the strain energy release rate,  $w$  the Dupré's energy of adhesion, and  $\Phi$  a dissipation function characteristic of the viscoelastic material and of the propagation in mode I, only depending on the temperature through the WLF shift factor  $a_0$  and on the crack propagation speed  $v$ . It is shown that the knowledge of the function  $\Phi$ , which represents viscoelastic losses localized at the crack tip, allows one to predict the kinetics of the spontaneous peeling and the force applied to the system. Experiments realized with two polyurethane strips in adhesive contact, one strip used as substrate being submitted to instantaneous or increasing tensile elongations, verify theoretical predictions with a reproducibility better than 3%.

## INTRODUCTION

Most varnishes, paints, and coatings have the drawback of flaking and peeling off, by propagating of a crack at the interface, when the substrate is deformed, for instance, by thermal expansion. In order to better understand this behavior, we have set up a simple model, together with an experimental arrangement consisting of two transparent rubber strips in adhesive contact, in which one strip is submitted to a fixed tensile elongation or to a fixed crosshead velocity.

This problem of the failure of lap shear joints has already investigated by Kendall,<sup>1</sup> using an energy balance theory based on optimizing the total energy of the system at the equilibrium. For instance, in a test at fixed load conditions, the equilibrium state is defined by  $dU_T = dU_E + dU_P + dU_S = 0$ , where  $U_T$ ,  $U_E$ ,  $U_P$ , and  $U_S$  are, respectively, total energy, stored elastic energy, potential energy of the load, and stored elastic energy at the interface, and the corresponding force is the adherence force. This concept has been successfully applied to a number of cases such as the adherence force of flat punches,<sup>2</sup> adherence force of spheres,<sup>3</sup> peeling,<sup>4</sup> interfacial failure in laminates<sup>5</sup> and composites.<sup>6</sup>

But the energy balance theory cannot give information about the stability of the system that depends upon the second derivative of the total energy. That is the reason why Maugis and Barquins<sup>7</sup> were led to reintroduce the concepts of fracture mechanics such as the strain energy release rate  $G$  and to study the stability according to the sign of the derivative of  $G$ . This approach has the advantage of enabling one to study the kinetics of crack propagation and to predict the evolution of the system whatever the geometry of contact and loading conditions.

The purpose of this paper is to show that the general equation of the adherence of elastomers  $G - w = w \cdot \Phi(a_\theta \cdot v)$ , where  $w$  is the Dupré's work of adhesion and  $\Phi$  a dissipation function, proposed by Maugis and Barquins in 1978 and verified for kinetics of adherence of spheres, of flat punches and peeling<sup>7-11</sup> also allows one to wholly describe the spontaneous peeling of rubber and to predict the time of complete delamination and the values of the corresponding force brought into play.

### KINETICS OF ADHERENCE

Lately, Maugis and Barquins<sup>12</sup> have shown that the contact of two elastic bodies can be treated as a thermodynamic problem. For instance, let us consider two elastic solids in contact over an area  $A$  under a load  $P$  corresponding to an elastic displacement  $\delta$ . The variation of the Helmholtz free energy of this system, neglecting thermal effect, is  $dF = P d\delta + (G - w) dA$ , where  $G$  is given by  $(\partial U_E / \partial A)_\delta$  and  $w$  is defined from the surface and interface energies of solids 1 and 2 by  $w = \gamma_1 + \gamma_2 - \gamma_{12}$ . Equilibrium at fixed grips conditions, corresponding to the maximum of  $F$ , is given by  $G = w$  (Griffith criterion). So, the relations giving the strain energy release rate and the elastic elongation as a function of the contact area and the force appear as the equations of state of the system. This equilibrium is stable if  $\partial G / \partial A$  is positive, unstable if negative, the elastic adherence force being the force corresponding to the particular case  $\partial G / \partial A = 0$ .

The equilibrium may be disrupted by a change in elongation. When  $G > w$ , the two bodies begin to separate and their separation can be seen as the propagation of a crack in mode I, the contact area decreasing as the crack advances. The difference  $(G - w)$  represents the crack extension force applied to the crack tip; it is the "motive" of the crack. Under this force, the crack takes a limiting speed  $v$  that depends on the temperature. If it is assumed that viscoelastic losses are proportional to  $w$ , as proposed by Gent and Schultz<sup>13</sup> and Andrews and Kinloch,<sup>14</sup> and are only localized at the crack tip, one can write<sup>7</sup>

$$G - w = w \cdot \Phi(a_\theta \cdot v) \quad (1)$$

where the second term is the viscous drag resulting from the losses limited to the crack tip.  $\Phi$  is a dimensionless function of the crack speed  $v$  and of the temperature through the shift factor  $a_\theta$  of the William-Landel-Ferry transformation.<sup>15</sup> This function  $\Phi$  is a characteristic of the viscoelastic material for the propagation in mode I, independent of the geometry of contact and the loading system. Knowledge of the function  $\Phi$  makes it possible to predict the evolution of the contact area in all circumstances. The prediction assumes only that rupture is adhesive, i.e., that the crack propagates at the interface and the application of eq. (1) implies that gross displacements are purely elastic, with  $G$  computed from the relaxed elastic modulus  $E_0$ , and that the frequency dependence of  $E$  only appears at the crack tip where deformation velocities are high. The loss factor  $E''/E'$  ( $E'$  and  $E''$  being the real and imaginary components of  $E$ ) and its frequency dependence are

taken into account in the function  $\Phi$ .<sup>16</sup> The interest of eq. (1) is that surface properties ( $w$ ) and viscoelastic losses ( $\Phi$ ) are clearly decoupled from the elastic properties, geometry, and loading conditions that only appear in  $G$ .

Adherence experiments carried out with a glass ball or a flat glass punch in contact with a polyurethane surface in push on-pull off tests, at fixed load,<sup>7</sup> cyclic load,<sup>9</sup> fixed displacement,<sup>10</sup> or fixed crosshead velocity,<sup>8</sup> verify theoretical predictions. Whatever the intrinsic properties (surface and viscoelastic) of tested solids or such experimental parameters (geometry of contact, speed of separation, temperature, relative humidity, initial applied force which presses together the two solids and duration of this initial contact), all experimental points fall on a master curve  $\Phi(v)$ . It is shown that over a large range of speeds of propagation ( $10^{-1} - 10^4 \mu\text{m/s}$ ), the function  $\Phi$  may be represented by the power function  $\Phi(\alpha_\theta \cdot v) = \alpha(\theta) \cdot v^n$ , where  $\alpha(\theta)$  is a parameter depending on the temperature<sup>8</sup> and  $n$  takes the value 0.6 for poly-urethane. Moreover, in eq. (1), the multiplicative effect of  $w$  on viscoelastic losses, can only arise if the interface is itself capable of withstanding stress,<sup>13</sup> has been confirmed by cylinder rolling experiments in an atmosphere of variable humidity.<sup>17,18</sup> One can point out that the model proposed by Greenwood and Johnson,<sup>19</sup> for a three-element viscoelastic material, gives a variation of  $G$  with the 0.5th power of the crack speed which is in reasonably good agreement with our experimental value ( $n = 0.6$ ).

In this paper, we show that eq. (1) enables one to study the kinetics of the spontaneous peeling of rubber, if a fixed tensile elongation or a fixed crosshead velocity is imposed on the elastic substrate.

### SPONTANEOUS PEELING

Let us consider [Fig. 1(a)] a long lap shear joint, similar to that investigated by Kendall,<sup>1</sup> made by contacting two smooth strips of elastomer, with the same Young modulus  $E$ , the same width  $b$ , and thickness  $h$ , in adhesive contact on to an area  $A = bL$ , without initial prestress. If an adequate tensile elongation  $\delta$  is imposed to the substrate, cracks appear at the free ends of the upper strip and propagate inwards along the interface [Fig. 1(b)], the spontaneous delamination being able to cause complete loss of contact. If, at a given time,  $x$  is the length of one relaxed peeled part of the upper strip, a simple application of Hooke's law to the double layer of length  $l$  and to the two strained free ends of the substrate allows one to calculate the force applied to the system by

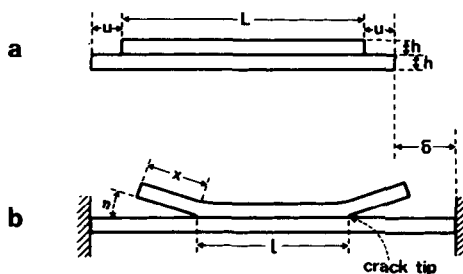


Fig. 1. Geometry of the first adhesive joint tested.

$$F = \frac{2bEH\delta}{L + 4u + 2x} \quad (2)$$

The corresponding stored elastic energy  $U_E = \frac{1}{2}F\delta$  is

$$U_E = \frac{bEh\delta^2}{L + 4u + 2x}$$

so that the strain energy release rate can be written as

$$G = Eh \left( \frac{\delta}{L + 4u + 2x} \right)^2 \quad (3)$$

Hence the equilibrium relationship is

$$\frac{\delta}{L + 4u + 2x} = \sqrt{\frac{w}{Eh}} \quad (4)$$

As  $(\partial G / \partial A)_\delta > 0$ , this equilibrium is stable, i.e., if a fluctuation decreases the contact area  $A$ ,  $G$  incrementally decreases, and one has  $G < w$ , the crack recedes to its equilibrium position. It can only advance if the tensile elongation  $\delta$  is varied bringing back  $G$  to the value  $w$ : one is dealing with the controlled rupture of an adhesive joint.

As soon as an instantaneous tensile elongation greater than  $\delta$  given by eq. (4), with  $x = 0$ , is applied to the substrate,  $G$  takes the corresponding value given by eq. (3) and immediately cracks are initiated at the extremities of the upper strip, and propagate in the interface [Fig. 1(b)]. Then,  $x$  increases and  $G$  continuously decreases with time. Figure 2 shows the relation between  $G$  and  $x$  for different instantaneous elongations in reduced coordinates, for the practical case  $u = 0$ . So, the minimum value of the uniaxial

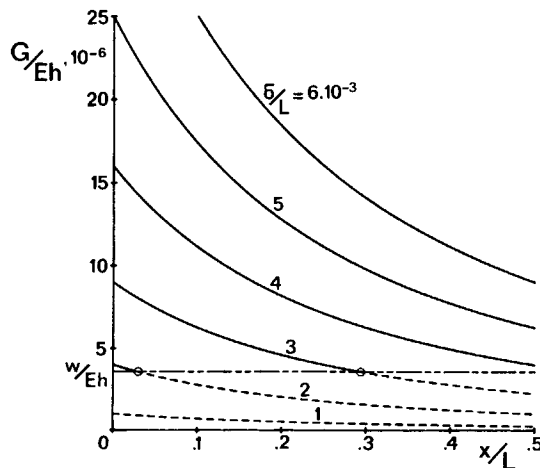


Fig. 2. Strain energy release rate versus length of the relaxed peeled strip, for various instantaneous tensile elongations, in reduced coordinates.

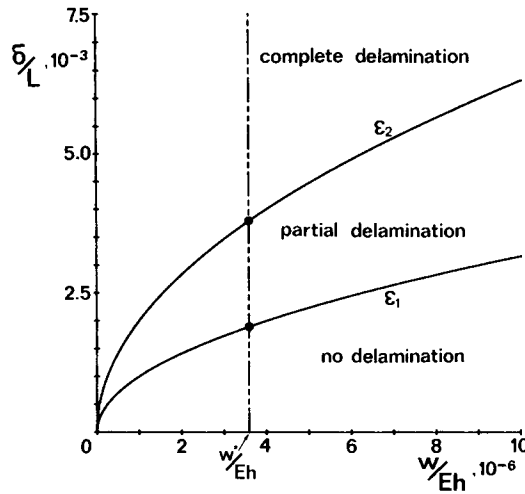


Fig. 3. Minimum and critical value of the instantaneous uniaxial deformation vs.  $w$ , to observe initiation of the crack and complete rupture of the contact, respectively, in reduced coordinates.

strain to observe the initiation of the crack can be deduced from eq. (4), and it is given by  $\epsilon_1 = \sqrt{w/Eh}$ .

Figure 2 allows one to see that the propagation can lead to an equilibrium state if  $G$  takes the value  $w$  before cracks reach the central part of the substrate, i.e.,  $x < L/2$ , or to the complete rupture of the contact if  $G > w$  with  $x = L/2$ . So, it is possible to define a new critical value of  $\delta$  corresponding to this borderline base. For the practical situation  $u = 0$ , one can deduce from eq. (4), with  $x = L/2$ , the minimum value of the instantaneous uniaxial deformation  $\epsilon_2 = \sqrt{4w/Eh}$  necessary to provoke the com-

TABLE I  
Geometries of the Adhesive Joints Tested, with Corresponding Formulae for Force, Strain Energy Release Rate and the Two Particular Values of the Instantaneous Uniaxial Deformation Applied to the Substrate

n°	GEOMETRY	F/Ebh	G/Eh	$\epsilon_1$	$\epsilon_2$
1		$\frac{25}{L+4u+2x}$	$\left(\frac{5}{L+4u+2x}\right)^2 = \frac{1}{4}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{w}{Eh}\right)^{\frac{1}{2}}$	$\left(\frac{4w}{Eh}\right)^{\frac{1}{2}}$
2		$\frac{25}{L+2u+x}$	$\left(\frac{5}{L+2u+x}\right)^2 = \frac{1}{4}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{w}{Eh}\right)^{\frac{1}{2}}$	$\left(\frac{9w}{Eh}\right)^{\frac{1}{2}}$
3		$\frac{36}{L+6u+4x}$	$\frac{3}{2}\left(\frac{5}{L+6u+4x}\right)^2 = \frac{1}{6}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{2w}{3Eh}\right)^{\frac{1}{2}}$	$\left(\frac{6w}{Eh}\right)^{\frac{1}{2}}$
4		$\frac{36}{L+3u+2x}$	$\frac{3}{2}\left(\frac{5}{L+3u+2x}\right)^2 = \frac{1}{6}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{2w}{3Eh}\right)^{\frac{1}{2}}$	$\left(\frac{6w}{Eh}\right)^{\frac{1}{2}}$
5		$\frac{36}{L+3u+x}$	$\frac{3}{4}\left(\frac{5}{L+3u+x}\right)^2 = \frac{1}{12}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{4w}{3Eh}\right)^{\frac{1}{2}}$	$\left(\frac{3w}{Eh}\right)^{\frac{1}{2}}$
6		$\frac{65}{2L+3u+x}$	$3\left(\frac{5}{2L+3u+x}\right)^2 = \frac{1}{12}\left(\frac{F}{Ebh}\right)^2$	$\left(\frac{4w}{3Eh}\right)^{\frac{1}{2}}$	$\left(\frac{3w}{Eh}\right)^{\frac{1}{2}}$

plete rupture of the contact. Figure 3 shows the variation of  $\epsilon_1$  and  $\epsilon_2$  with the thermodynamic work of adhesion  $w$ , in reduced coordinates.

Similar considerations can be formulated for other types of adhesive joints. These models, experimentally tested, are described in Table I, with the corresponding formulae for the strain energy release rates, forces, and critical instantaneous deformation values  $\epsilon_1$  and  $\epsilon_2$ .

The application of Hooke's law to the system also enables one to write the relation including all the geometrical parameters, so that the length  $x$  of each peeled strip is given by the positive root of the equation:

$$4x^2 + 2x(l + 4u + \delta) - L(L + 4u + \delta - l) + 4ul = 0 \quad (5)$$

of which we can deduce the crack speed at fixed elongation  $\delta$ ,

$$\frac{dx}{dt} = -\frac{1}{2} \cdot \frac{L + 4u + 2x}{l + 4u + 4x + \delta} \cdot \frac{dl}{dt} \quad (6)$$

and at fixed crosshead velocity  $\dot{\delta}$ ,

$$\frac{dx}{dt} = \frac{-(dl/dt)(L + 4u + 2x) + \dot{\delta}(L - 2x)}{2(l + 4u + 4x + \delta)} \quad (7)$$

the variation with time of the length  $l$  of the adhesive part of the joint,  $dl/dt$ , being determined by the slope in every point of experimental curves  $l(t)$ .

So, the strain energy release rate, given by eq. (3), with  $x$  deduced from eq. (5), can be related to the crack speed  $dx/dt$  assessed by eq. (6) or (7) following the experimental procedure, in order to verify eq. (1), previously proposed and experienced in normal approach measurements of adherence and peeling.<sup>7-12,18</sup>

## EXPERIMENTAL METHOD

The viscoelastic material chosen was an optically smooth polyurethane, recommended for dynamic studies in photoelasticity (PSM4 Vishay, with  $E = 3.6$  MPa), similar to that used for previous experiments on the kinetics of adherence of glass spheres, or glass flat punches and for peeling tests.<sup>7</sup> It was delivered as plates of thicknesses  $h = 3.175$  mm and 6.350 mm. The surfaces were wiped with an alcohol-soaked rag, dried with warm air and left, sheltered from dust, for 30 min for the equilibrium with room temperature to be reached. Then, two strips with various lengths  $L$  and widths  $b$  varying in the range 5–20 cm and 5–20 mm, respectively, were gently superimposed, and they adhered under the only molecular attraction forces, without additional adhesive. In order to avoid the dwell time effect,<sup>20</sup> strips were coupled during the same contact duration 30 min, for any set of experiment. Moreover, temperature (23°C) and humidity (84%) were kept constant. In these conditions, the reproducibility is better than 3%.

Experiments at fixed grips conditions and at fixed crosshead velocity were carried out using a tensile machine (Instron 1026) that enables one to im-

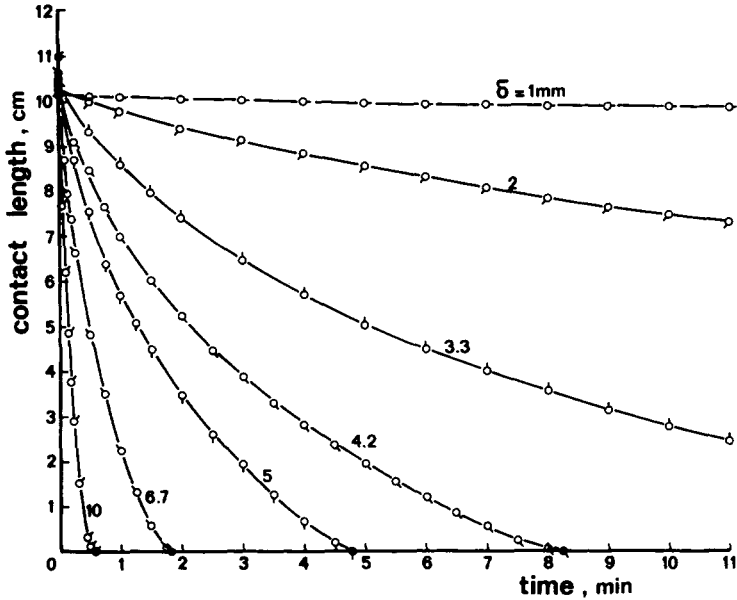


Fig. 4. Variation with time of the contact length between two strips of polyurethane, for various instantaneous tensile elongations applied to the substrate.  $L = 100$  mm,  $b = 10$  mm,  $h = 3.175$  mm,  $u = 1$  mm.

pose, with help of two gear boxes, speeds  $\delta$  varying in the range 0.5–500 mm/min, and to measure forces up to 50 N. For a quantitative evaluation of the crack speed during the delamination, a 16-mm camera recorded the experimental arrangement at a rate varying from 10 to 50 frames/s; the frames were then enlarged and the distance  $l$  between the two crack tips were measured, and the peel angle  $\eta$ , also [Fig. 1(b)].

## EXPERIMENTAL RESULTS AND DISCUSSION

In a first set of experiments, we have studied the kinetics of spontaneous peeling at fixed tensile elongation. The time necessary to impose this constant elongation was in any case inferior to 1 s, a very short duration with respect to the rupture time. Figure 4 shows the variation with time of the distance  $l$  between the two crack tips (contact length) for different "instantaneous" tensile elongations  $\delta$  applied to a strip 100 mm long, 10 mm wide, 3.175 mm thick, and with  $u = 1$  mm. Experiments were stopped after 11 min due to the limited film capacity of the camera.

The important and fortunate observation, for a good validity of the model, was that, at a given time, the two cracks exactly propagate with the same speed, i.e., that the experimental arrangement remained symmetrical up to the complete rupture of contact, when it was observed.

Corresponding values of the strain energy release rate and the associated crack speed, calculated by eq. (3) and (6), respectively, are represented on Figure 5 on log-log coordinates. All the points fall on the same straight line that corroborates the previous findings<sup>7</sup> and consequently confirms eq. (1) in which the function  $\Phi$  for polyurethane samples varies as the 0.6th power of the crack speed. Taking into account  $\alpha(\theta) = 4.75 \times 10^4$  SI units,<sup>8</sup>

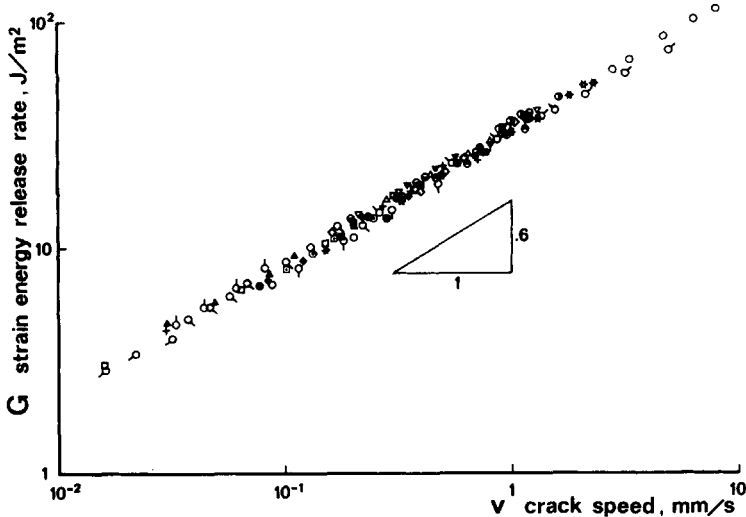


Fig. 5. Strain energy release rate versus crack speed for strips of polyurethane in adhesive contact, for various instantaneous tensile elongations and cross head velocities.

corresponding to the room temperature 23°C, and data from Figure 5, the mean value of Dupré's work of adhesion that can be calculated by eq. (1) is  $w = 41 \text{ mJ/m}^2 (\pm 1 \text{ mJ/m}^2)$ , a fairly low value due to the high humidity ratio.<sup>17,18</sup> This value is used as reference  $w^*$  on Figures 2 and 3 and corresponds to  $\epsilon_1 \simeq 1.9 \times 10^{-3}$  and  $\epsilon_2 \simeq 3.8 \times 10^{-3}$ .

Figure 6 shows variations recorded by the tensile machine of the force to which the experimental arrangement is subjected with time, in the same conditions as in Figure 4. As expected, an increasing in tensile elongation provokes an proportional increasing in the initial force, and the time required to observe complete delamination is shorter, the force then remaining constant.

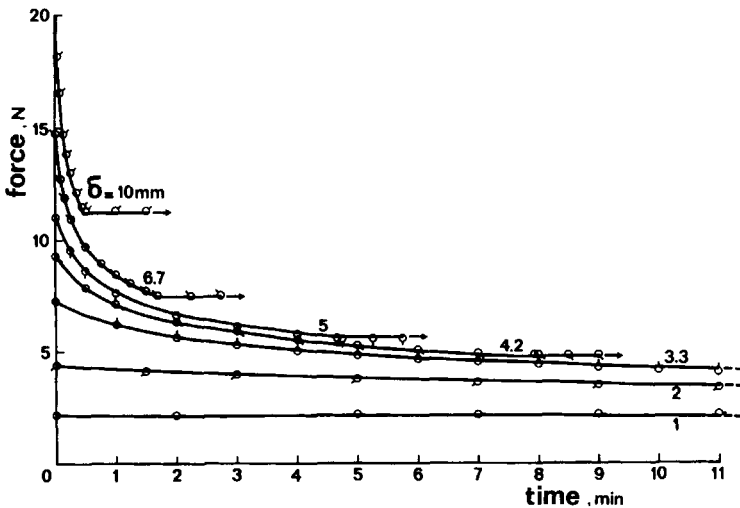


Fig. 6. Variation with time of the force applied to the system in the same conditions as in Figure 4. Experimental results and corresponding computed curves (heavy lines):  $L = 100 \text{ mm}$ ,  $b = 10 \text{ mm}$ ,  $h = 3.175 \text{ mm}$ ,  $u = 1 \text{ mm}$ .



By numerical integration of the general equation for the adherence of viscoelastic materials [eq. (1)], taking into account eqs. (2) and (3) and the variation of  $\bar{\phi}$  with the 0.6th power of the crack speed, it is possible to predict variations of the force during the spontaneous peeling. The computed curves (heavy lines of Fig. 6) are in quite good agreement with experimental results. It can be pointed out that the small vertical shift visible on some curves arises from a weak variation of  $w$  with respect to the mean value used for calculations. Moreover, the light divergences observed for long times are probably due to dwell time effect resulting from the relaxation of stresses stored in the roughnesses at the interface.<sup>20</sup>

In a second set of experiments, the lower strip of polyurethane used as elastic substrate, was submitted to various fixed crosshead velocities. Variations of the contact length  $l$  vs. time are shown in Figure 7. The kinetics of separation is less easy to interpret as at fixed tensile elongation due to the competition between increasing elongation  $\delta$  at constant contact length  $l$  and decreasing  $l$  at constant  $\delta$  for increasing  $G$ :

$$\frac{dG}{dt} = \left(\frac{\partial G}{\partial \delta}\right)_l \cdot \frac{d\delta}{dt} + \left(\frac{\partial G}{\partial l}\right)_\delta \cdot \frac{dl}{dt}$$

At the beginning, as long as the strain energy release rate remains smaller than the thermodynamic work of adhesion  $w$ , the cracks cannot start and obviously the contact length  $l$  increases with  $\delta$ . Moreover, when  $G$  just becomes slightly greater than  $w$ , the cracks begin to propagate with a very slow speed so that the contact length increases for a short time and then decreases until complete delamination is reached.

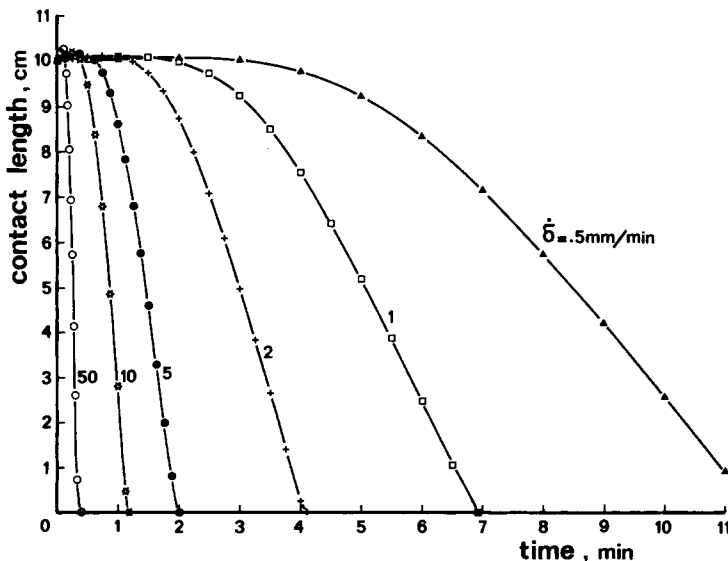


Fig. 7. Variation with time of the contact length between two strips of polyurethane, for various crosshead velocities imposed to the substrate:  $L = 100$  mm,  $b = 10$  mm,  $h = 3.175$  mm,  $u = 1$  mm.

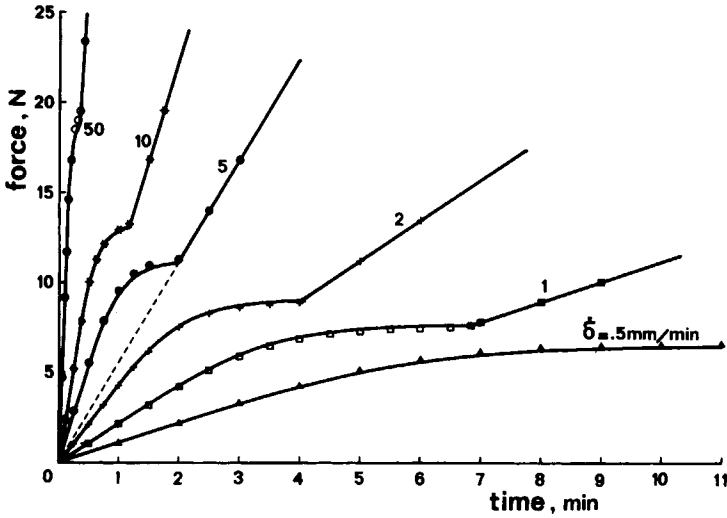


Fig. 8. Variation with time of the force applied to the system in the same conditions as in Figure 7. Experimental results and corresponding computed curves (heavy lines):  $L = 100$  mm,  $b = 10$  mm,  $h = 3.175$  mm,  $u = 1$  mm.

Corresponding recorded forces are shown on Figure 8. Due to the lower variation of the crack speed just before the total rupture of the contact, the force is seen to increase less rapidly. Thereafter, as expected, the force increases linearly with time and corresponds to the continuous elongation of the substrate. Computed curves (heavy lines) obtained by numerical integration of eq. (1) and using eq. (2) and (7) are in good agreement with experimental results.

Relations between the strain energy release rate and the associated crack speed are given in Figure 5. They wholly confirm the variation of the function  $\Phi$  with the 0.6th power of the propagation speed.

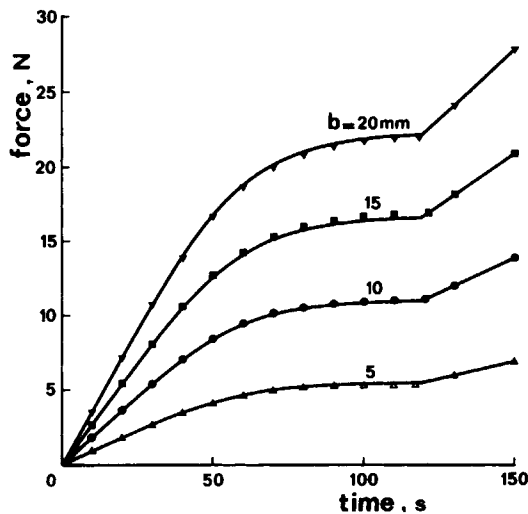


Fig. 9. Force versus time, for various contact widths. Experimental results and corresponding computed curves (heavy lines);  $\delta = 5$  mm/min,  $L = 100$  mm,  $h = 3.175$  mm,  $u = 1$  mm.

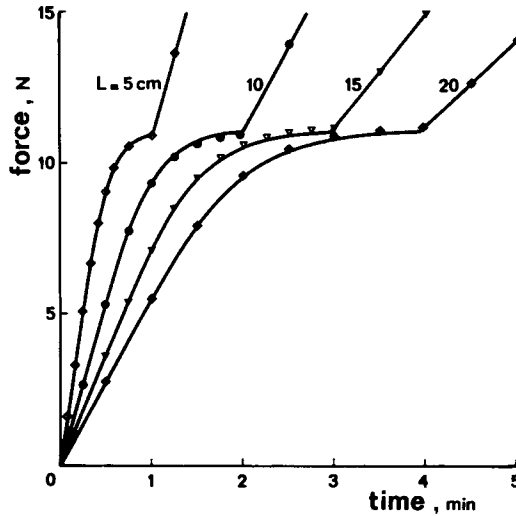


Fig. 10. Force versus time, for various initial contact lengths. Experimental results and corresponding computed curves (heavy lines):  $\delta = 5$  mm/min,  $b = 10$  mm,  $h = 3.175$  mm,  $u = 1$  m.

In order to further verify the good validity of the model, influences of the initial contact length  $L$ , of the width  $b$ , and of the free end length  $u$  of the elastic substrate have been studied, when the latter is submitted to the same fixed crosshead velocity  $\delta = 5$  mm/min. Experimental results and corresponding computed curves are compared in Figures 9, 10, and 11. As expected, a change in contact width  $b$ , all other geometrical parameters remaining constant, does not alter the kinetics of propagation; indeed, the curves  $l(t)$  and hence  $x(t)$  are all superimposed, so that at every time the recorded force is proportional to the width  $b$  [eq. (2)] as shown on the Figure

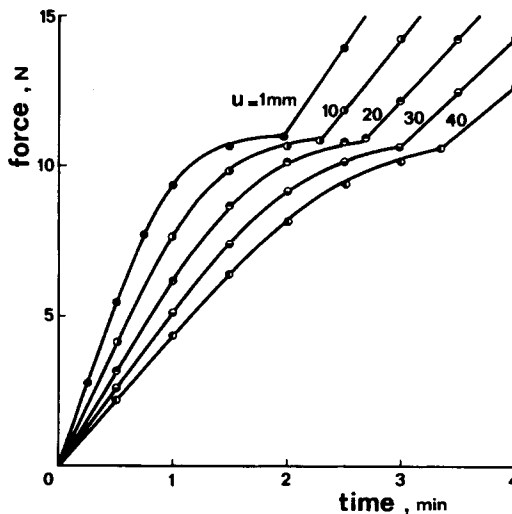


Fig. 11. Force versus time, for various initial lengths of the free ends of the substrate. Experimental results and corresponding computed curves (heavy lines):  $\delta = 5$  mm/min,  $L = 100$  mm,  $h = 3.175$  mm,  $b = 10$  mm.

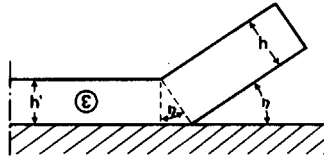


Fig. 12. Schematical geometry of the peeled strip in the vicinity of the crack tip.

9, and the complete delamination is observed after the same duration. On the other hand, an increase in the initial contact length  $L$  delays the appearance of the total rupture of contact, and if  $u$  can be neglected with respect to  $L$ , the corresponding force is constant, as shown in Figure 10, and the time to observe complete rupture is proportional to the initial contact length. Lastly, Figure 11 confirms that the length of the free ends of the substrate plays a considerable part in the kinetics of propagation but only a small part in the force applied to the system.

During crack propagation, the spontaneous peeling angle was measured, and a slow increase with time was observed. Simple geometrical considerations associated with the incompressible character of the tested material enable one to relate the value of angle to the uniaxial strain in the adhering upper strip (Fig. 12). Indeed, to a first approximation, the flaking of the peeled strip can be easily ascribed to the variation of the thickness between the two adhering and relaxed states, and we can write  $\cos \eta = h'/h = 1 - \epsilon/2$ , hence, if  $\epsilon$  is not too high,  $\eta \simeq \sqrt{\epsilon}$ . Although, the substrate is not a rigid body, and is not perfectly plane in the vicinity of contact zone due to the asymmetry of the experimental arrangement, measurements of peeling angle confirm the prediction with an accuracy of better than 8%.

The other types of models described in Table I have been tested at fixed tensile elongation as at fixed crosshead velocity. For instance, Figure 13 shows the fifth geometry of Table I during crack propagation. This figure proves that the system remains perfectly symmetrical until the complete rupture without perceivable influence on peeling angles of weights of already detached portions. Corresponding relations between strain energy release rate and crack speed are given in Figure 5. As expected, all the results confirm previous findings whatever the geometry of the adhesive joint. So it is proved that the knowledge of the function  $\Phi$  allows one to predict the evolution of the system, among other things, the force involved and contact duration before rupture. For instance, Figure 14 shows the good agreement between experimental results and theoretical curves (heavy lines) obtained with the six arrangements studied in a same test at fixed crosshead velocity.

So we think that, due to the elementary nature of the model, and also its sound validity, it may be suitable for solving certain practical problems such as the ability of varnishes, paints, and coatings to adhere on deformable substrates.

## CONCLUSION

The concepts of fracture mechanics may be used to study the kinetics of the spontaneous peeling of a rubber strip in adhesive contact with an elastic substrate, using the general equation

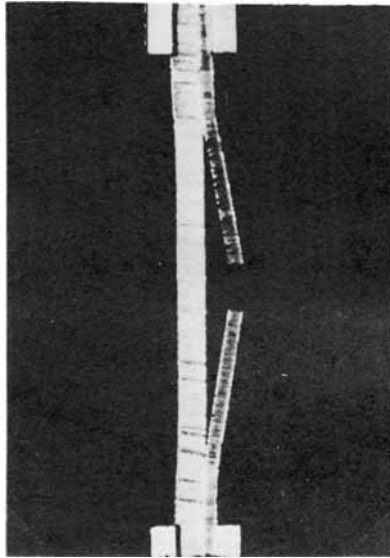


Fig. 13. View of the fifth geometry during crack propagation ( $L = 100$  mm,  $b = 10$  mm,  $h = 3.175$  mm,  $u = 0$ ,  $\delta = 5$  mm/min)

$$G - w = w \cdot \Phi(a_\theta \cdot v)$$

where  $G$  is the strain energy release rate,  $w$  is Dupré's work of adhesion, and  $\Phi$  is a dissipation function characteristic of the viscoelastic material for the crack propagation in mode I. This function  $\Phi$  depends only on the temperature, through the term  $a_\theta$  (William-Landel-Ferry factor) and on

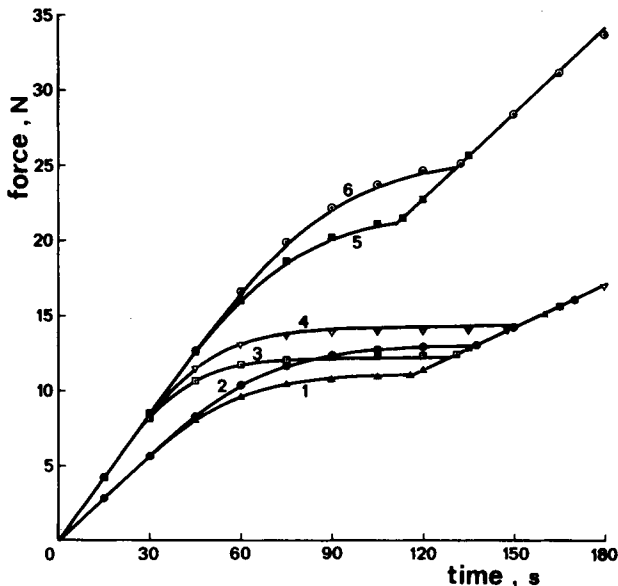


Fig. 14. Comparison for the same crosshead velocity between the six geometries tested. Experimental results and corresponding computed curves (heavy lines):  $\delta = 5$  mm/min,  $L = 100$  mm,  $h = 3.175$  mm,  $b = 10$  mm,  $u = 0$ .

the speed  $v$  of the crack propagating at the interface, and is consequently independent of the geometry and of the type of test. Experiments carried out with various arrangements at fixed tensile elongations and at fixed crosshead velocities show that, over a large range of speeds of propagation, the function  $\Phi$  may be represented by the power function  $\Phi(a_\theta \cdot v) = a(\theta) \cdot v^n$ , with  $n = 0.6$  for polyurethane tested. This result corroborates previous findings concerning the kinetics of adherence between a spherical or flat rigid punch and the same rubberlike material.

Knowledge of the function  $\Phi$  thus makes it possible to predict the kinetics of propagation in all particular cases provided that the elongations remain purely elastic (with viscoelastic losses being left localized at the crack tip) and that the rupture is the rupture of an adhesive joint (with propagation of the crack in the interface). Particularly, the differential equation above may be used to predict: the time of spontaneous delamination as a function of fixed tensile elongation or crosshead velocity, the size of the contact area and the force brought into play at any time during peeling. Experimental results verify theoretical predictions with an accuracy better than 3%.

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### References

1. K. Kendall, *J. Phys. D: Appl. Phys.*, **8**, 512 (1975).
2. K. Kendall, *J. Phys. D: Appl. Phys.*, **4**, 1186 (1971).
3. K. L. Johnson, K. Kendall, and A. D. Roberts, *Proc. Roy. Soc. Lond.* **A324**, 301 (1971).
4. K. Kendall, *J. Phys. D: Appl. Phys.*, **5**, 1782 (1973).
5. K. Kendall, *J. Phys. D: Appl. Phys.*, **8**, 1722 (1975).
6. K. Kendall, *Proc. Roy. Soc. Lond.*, **A341**, 409 (1975).
7. D. Maugis and M. Barquins, *J. Phys. D: Appl. Phys.*, **11**, 1989 (1978).
8. M. Barquins and D. Maugis, *J. Adhesion*, **13**, 53 (1981).
9. M. Barquins, *Int. J. Adhesion Adhesives*, **3**, 71 (1983).
10. M. Barquins, *J. Appl. Polym. Sci.*, **28**, 2647 (1983).
11. D. Maugis and M. Barquins, *J. Phys. D: Appl. Phys.*, **16**, 1843 (1983).
12. D. Maugis and M. Barquins, in *Adhesion and Adsorption of Polymers*, L. H. Lee, Ed., Plenum, New-York, 1980, Vol. 12A, pp. 203-277.
13. A. N. Gent and J. Schultz, *J. Adhesion*, **3**, 281 (1972).
14. E. H. Andrews and A. J. Kinloch, *Proc. Roy. Soc. Lond.*, **A332**, 385 (1973).
15. J. D. Ferry, *Viscoelastic Properties of Polymers*, 2nd ed., Wiley, New York, 1970, Chap. 10.
16. D. Maugis, in *Microscopic Aspects of Adhesion and Lubrication*, Tribology Series 7, J. M. Georges, Ed., Elsevier, Amsterdam, 1982, pp. 221-252.
17. A. D. Roberts, *Rubber Chem. Technol.*, **32**, 23 (1979).
18. M. Barquins, thesis, University of Paris, 1980.
19. J. A. Greenwood and K. L. Johnson, *Phil. Mag.*, **A43**, 697 (1981).
20. M. Barquins, *J. Adhesion*, **14**, 63 (1982).

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