

WEAK ADHESIVE JUNCTIONS IN THE PRESENCE OF INTERMOLECULAR INTERACTIONS

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Polymer adhesive has been the subject of many theoretical and experimental studies in the last few years [1]. Certain weak adhesive junctions [2] may be described by a mechanical model where: (i) the opening rate dh/dt vanishes when the stress σ is below a certain threshold value σ^* and (ii) for $\sigma > \sigma^*$, the opening rate increases with the stress as $dh/dt = Q^{-1}(\sigma - \sigma^*)$ where Q is a friction coefficient. This model, called in the literature the Model II, has been explored in detail [3][4]. Intermolecular interactions, which give rise to the thermodynamic work of adhesive W , have been recently incorporated into this model in the limit of zero crack velocity [5][6]. In this paper we generalise these studies to finite crack velocities. In particular, we calculate the length of the junction L as well as the fracture energy G .

Consider the problem of a mode I semi-infinite crack embedded in a linear purely elastic material. For plane strain conditions the normal stress distribution $\sigma(x)$ and the crack displacement $u(x)$ are respectively given by [7]

$$\sigma(x) = \frac{K}{\sqrt{2\pi x}} \quad x > 0; \quad \sigma(x) = 0 \quad x < 0 \quad (1)$$

and

$$u(x) = \frac{2(1-\nu)}{\mu} K \sqrt{\frac{-x}{2\pi}} \quad x < 0; \quad u(x) = 0 \quad x > 0 \quad (2)$$

where μ is the shear modulus, ν the Poisson ratio and K the stress intensity factor. The fracture energy per unit area G is given by Irwin's equation [8]

$$G = \frac{1-\nu}{2\mu} K^2. \quad (3)$$

In a cohesive zone model the singular stress (1) is relaxed by inelastic deformation in a zone directly ahead of the crack (see Fig. 1). In this region ($0 < x < L$) the elastic field may be described in terms of a source function $\Phi(x)$ defined by [9][3]

$$\sigma(x) = \mu \int_0^x dy \Phi(y) (x-y)^{-1/2} \quad (4)$$

and

$$u(x) = 2(1-\nu) \int_x^L dy \Phi(y) (y-x)^{1/2}. \quad (5)$$

It can be shown [10] that (4) and (5) are equivalent to the standard formulation [11]

$$u(x) = \frac{2(1-\nu)}{\mu} K \sqrt{\frac{L-x}{2\pi}} - \frac{1-\nu}{\pi\mu} \int_0^L dy \sigma(y) \ln \left| \frac{\sqrt{L-x} + \sqrt{L-y}}{\sqrt{L-x} - \sqrt{L-y}} \right|. \quad (6)$$

At large distance ($|x| \gg L$), (4) and (5) reduce to (1) and (2) with

$$\frac{K}{\sqrt{2\pi\mu}} = \int_0^L dy \Phi(y). \quad (7)$$

The mechanical behaviour of the junction is described [5][12] by the following law relating the rate of opening $dh/dt=2du/dt$ and the normal stress σ (Model II)

$$\frac{dh}{dt} = Q^{-1}(\sigma - \sigma^*) \quad \sigma > \sigma^*; \quad \frac{dh}{dt} = 0 \quad \sigma < \sigma^*. \quad (8)$$

where Q is the friction coefficient of the junction and σ^* is a threshold stress. Note that the opening rate is continuous at $\sigma = \sigma^*$.

We now consider the problem of a steadily growing crack with translational velocity V . Inserting (4) and (5) into (8) we obtain the fundamental equation

$$\Delta_\lambda \Phi(x) = \frac{\sigma^*}{\mu} \quad (9)$$

where Δ_λ is the following linear integral operator

$$\Delta_\lambda \Phi(x) \equiv \int_0^x dy \Phi(y) (x-y)^{-1/2} - \lambda \int_x^L dy \Phi(y) (y-x)^{-1/2} \quad (10)$$

and

$$\lambda = V/V^* \text{ a reduced velocity with } V^* = \mu/[2(1-\nu)Q].$$

Equation (9) is a non-homogeneous linear integral equation for the source function $\Phi(x)$. It must be supplemented by the boundary condition

$$h_f = 2u(x=0) \quad (11)$$

where h_f is the terminal value of the junction opening. Using (5), (11) can be rewritten as

$$\frac{h_f}{4(1-\nu)} = \int_0^L dy \Phi(y) y^{1/2}. \quad (12)$$

We have also the conditions that

$$(i) \text{ in the absence of connectors } G \text{ reduces to } W, \quad (13)$$

$$(ii) \text{ in the zero-rate limit } (V \rightarrow 0) G \text{ reduces to } G_0 = W + \sigma^* h_f \text{ [6]}. \quad (14)$$

The solution of (9) can be written as

$$\Phi(x) = \Phi_H(x) + \Phi_M(x) \quad (15)$$

where $\Phi_H(x)$ and $\Phi_M(x)$ satisfy respectively

$$\Delta_\lambda \Phi_H(x) = \frac{\sigma^*}{\mu} \quad (16)$$

and

$$\Delta_\lambda \Phi_M(x) = 0. \quad (17)$$

The source function $\Phi_H(x)$ was derived by Fager et al. [4],

$$\Phi_H(x) = \frac{\sigma^*}{\mu} \frac{1}{B(\frac{1}{2}-\epsilon, \frac{1}{2}+\epsilon)} x^{-\frac{1}{2}+\epsilon} (L-x)^\epsilon \quad (18)$$

where ϵ is related to the reduced velocity λ by

$$\tan(\pi\epsilon) = \lambda. \quad (19)$$

In (18), $B(x,y)$ denotes the beta function $B(x,y) = \Gamma(x) \Gamma(y) / \Gamma(x+y)$ where $\Gamma(x)$ is the gamma function [13]. The source function $\Phi_M(x)$, which originates from the intermolecular forces, is given by

$$\Phi_M(x) = \left(\frac{W}{\pi\mu(1-\nu)} \right)^{1/2} \frac{1}{B(\frac{1}{2}-\epsilon, \epsilon)} L^{1/2} x^{-\frac{1}{2}+\epsilon} (L-x)^{-1+\epsilon}. \quad (20)$$

This form is obtained by solving (18) with the conditions (14) and (15). The dependence

$$\Phi_M(x) \propto x^{-\left(\frac{1}{2}+\varepsilon\right)}(L-x)^{-1+\varepsilon}$$

was already proposed by de Gennes and Troian in their investigation of the so-called Model I [14]. Knowing $\Phi(x) = \Phi_H(x) + \Phi_M(x)$, we can now calculate the length of the junction L , see (12), the stress intensity factor K , see (7) and finally the fracture energy G , see (3).

The length L of the cohesive zone can be determined by using the boundary condition (11). Inserting (15) into (12) we obtain

$$\frac{h_f}{4(1-\nu)} = \frac{\sigma^* B(1-\varepsilon, 1+\varepsilon)}{\mu B\left(\frac{1}{2}-\varepsilon, \frac{1}{2}+\varepsilon\right)} L + \left(\frac{W}{\pi\mu(1-\nu)}\right)^{1/2} \frac{B(1-\varepsilon, \varepsilon)}{B\left(\frac{1}{2}-\varepsilon, \varepsilon\right)} L^{1/2}. \quad (21)$$

Hence

$$\frac{L}{L_0} = \left[\frac{B\left(\frac{1}{2}, \frac{1}{2}-\varepsilon\right) \sqrt{1 + \frac{\sigma^* h_f}{W} + 1}}{B\left(\frac{1}{2}, \frac{1}{2}\right) \sqrt{1 + \frac{\sigma^* h_f B\left(1+\varepsilon, \frac{1}{2}-\varepsilon\right)}{W B\left(1-\varepsilon, \frac{1}{2}+\varepsilon\right)} + 1}} \right]^2 \quad (22)$$

where L_0 denotes the zero-rate limit

$$L_0 = \frac{\pi \mu h_f^2}{4(1-\nu) W} \frac{1}{\left(\sqrt{1 + \frac{\sigma^* h_f}{W} + 1}\right)^2}. \quad (23)$$

At low velocities ($V \ll V^*$) we obtain

$$L = L_0 \left(1 + \frac{4 \ln 2}{\sqrt{1 + \frac{\sigma^* h_f}{W}}} \frac{V}{V^*} + \dots \right) \quad \frac{V}{V^*} \ll 1. \quad (24)$$

At higher velocities ($V \gg V^*$) two cases must be considered depending on the value of the parameter $\sigma^* h_f / W$

(i) if $\sigma^* h_f / W \gg 1$ then

$$L \cong \frac{1}{2(1-\nu)} \frac{\mu h_f V}{\sigma^* V^*} \quad \frac{V}{V^*} \gg 1 \quad (25)$$

(ii) if $\sigma^*h_f/W \ll 1$ then

$$L \equiv \frac{\pi}{16(1-\nu)} \frac{\mu h_f^2}{W} \left(\frac{V}{V^*} \right)^2 \quad 1 \ll \frac{V}{V^*} \ll \frac{W}{\sigma^*h_f} \quad (26)$$

$$L \equiv \frac{1}{2(1-\nu)} \frac{\mu h_f}{\sigma^*} \frac{V}{V^*} \quad 1 \ll \frac{W}{\sigma^*h_f} \ll \frac{V}{V^*}$$

Figure 2 represents the behaviour of L/L_0 as a function of the reduced velocity V/V^* for three values of the parameter σ^*h_f/W .

The stress intensity factor K can be determined by using (7). Inserting (15) into (7) we have

$$\frac{K}{\sqrt{2\pi\mu}} = \frac{\sigma^*B(\frac{1}{2}-\epsilon, 1+\epsilon)}{\mu B(\frac{1}{2}-\epsilon, \frac{1}{2}+\epsilon)} L^{1/2} + \left(\frac{W}{\pi\mu(1-\nu)} \right)^{1/2}. \quad (27)$$

Substitution of (22) into (27) yields

$$K^2 = \frac{2\mu}{1-\nu} \left[W + \sigma^*h_f \frac{B(1+\epsilon, \frac{1}{2}-\epsilon)}{B(1-\epsilon, \frac{1}{2}+\epsilon)} \right]. \quad (28)$$

Using Irwin's equation (3) we obtain the fracture energy of the junction

$$G = W + \sigma^*h_f \frac{B(1+\epsilon, \frac{1}{2}-\epsilon)}{B(1-\epsilon, \frac{1}{2}+\epsilon)} \quad (29)$$

with the limiting behaviours

$$G = G_0 \left(1 + \frac{4\ln 2}{\pi} \frac{1}{1 + \frac{W}{\sigma^*h_f} \frac{V}{V^*}} + \dots \right) \quad \frac{V}{V^*} \ll 1 \quad (30)$$

$$G \equiv \sigma^*h_f \frac{\pi}{2} \frac{V}{V^*} \quad \frac{V}{V^*} \gg 1$$

where G_0 is the zero-rate limit: $G_0 = W + \sigma^*h_f$ [6].

Figure 3 represents the behaviour of $(G-W)/G_0-W$ as a function of the reduced velocity V/V^* . This function is independent of the parameter σ^*h_f/W , see (29). Equation (29) indicates that at any velocity the fracture energy is a

linear combination of the thermodynamic work of adhesion and the pull-out work.

In this paper we have investigated the properties of the so-called Model II taking into account molecular interactions. This model may be used to describe the process of chain pull-out and the relation between chain pull-out and interface toughness in the adhesion of elastomers [15][16]. Our main results are expressions (22) and (29) for the length L of the junction and for the fracture energy G as a function of the reduced velocity V/V^* , the thermodynamic work of adhesion W and the zero-rate limit of the pull-out work σ^*h_r .

Note that in our model the stress $\sigma(x)$ ahead of the cohesive zone tip becomes much larger than σ^* as one approaches $x = L$. Nevertheless, the junction remains closed for $x > L$ for one must first reach a sufficiently large stress to overcome the attractive intermolecular forces and to separate the two sides of the junction. Only then can the opening process described by (8) take place.

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REFERENCES

- [1] H.R. Brown, *Annual Review of Material Science* 21 (1991) 463.
- [2] P.-G. de Gennes, *Canadian Journal of Physics* 68 (1990) 1049.
- [3] P.-G. de Gennes, *Journal of Physics, France* 5 (1989) 2551; *Comptes Rendus Academie des Sciences Paris*, t. 309, Série II (1989) 1125.
- [4] L.-O. Fager, J.L. Bassani, C.-Y. Hui and D.-B. Xu, *International Journal of Fracture Mechanics* 52 (1991) 119.
- [5] E. Raphaël and P.-G. de Gennes, *Journal of Physical Chemistry* 96 (1992) 4002.
- [6] H.R. Brown, C.-Y. Hui and E. Raphaël, *Macromolecules* 27 (1994) 608.
- [7] M.F. Kanninen and C.H. Popelar, *Advanced Fracture Mechanics*, Oxford University Press (1985).
- [8] G.R. Irwin, *Applied Materials Research* 3 (1964) 65.
- [9] A. Cottrell, in *Physics of Strength and Plasticity*, A.S. Argon (ed.), MIT Press (1969) 257.
- [10] C.-Y. Hui and E. Raphaël, *International Journal of Fracture Mechanics* 61 (1993) R51.

[11] H. Tada, P.C. Paris and G.R. Irwin, *Stress Analysis of Cracks Handbook*, Del Research Corporation, Hellertown, PA (1973).

[12] E. Raphaël and P.-G. de Gennes, *Les Houches, Soft Order in Physical Systems*, R. Bruinsma and Y. Rabin (eds.) (1994).

[13] I.S. Gradshteyn and I.M. Ryzhik, *Tables of Integrals, Series and Products*, A. Jeffery (ed.) Academic Press, New York (1965) 933.

[14] P.-G. de Gennes and S.M. Troian, *Comptes Rendus Academie des Sciences Paris*, t. 311, Série II (1990) 389.

[15] H.R. Brown, *Macromolecules* 26 (1993) 1666.

[16] C. Creton, H.R. Brown and K.R. Shull, *Macromolecules*, submitted.

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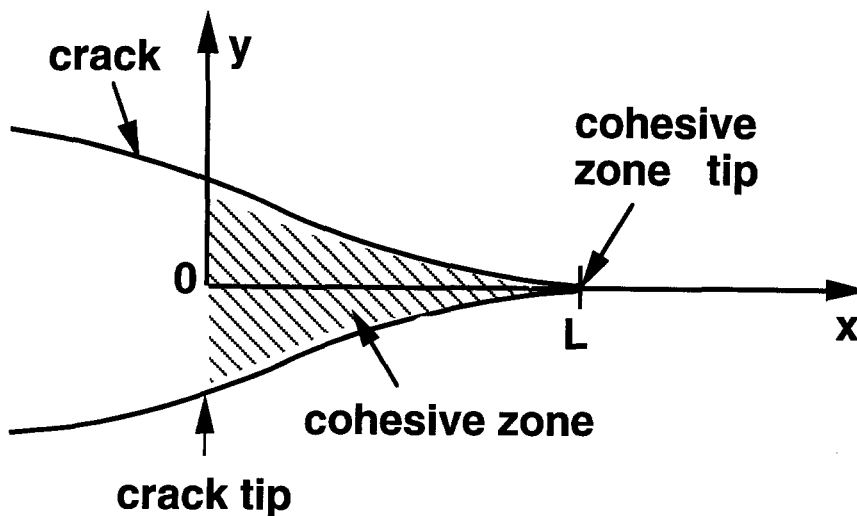


Figure 1. A global view of the cohesive zone.

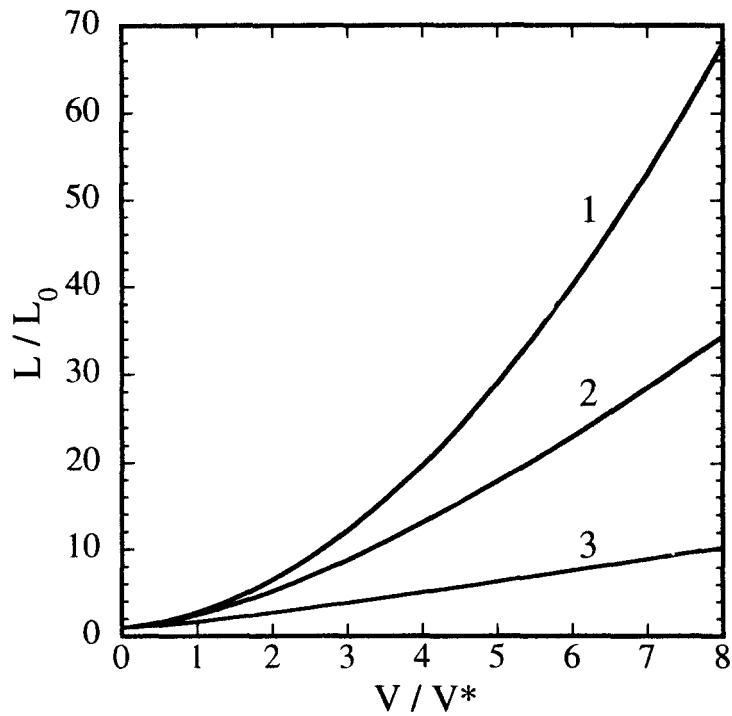


Figure 2. Plots of L/L_0 versus V/V^* for different values of the parameter σ^*h_f/W : curve 1, $\sigma^*h_f/W=0.01$; curve 2, $\sigma^*h_f/W=0.25$; curve 3, $\sigma^*h_f/W=5$.

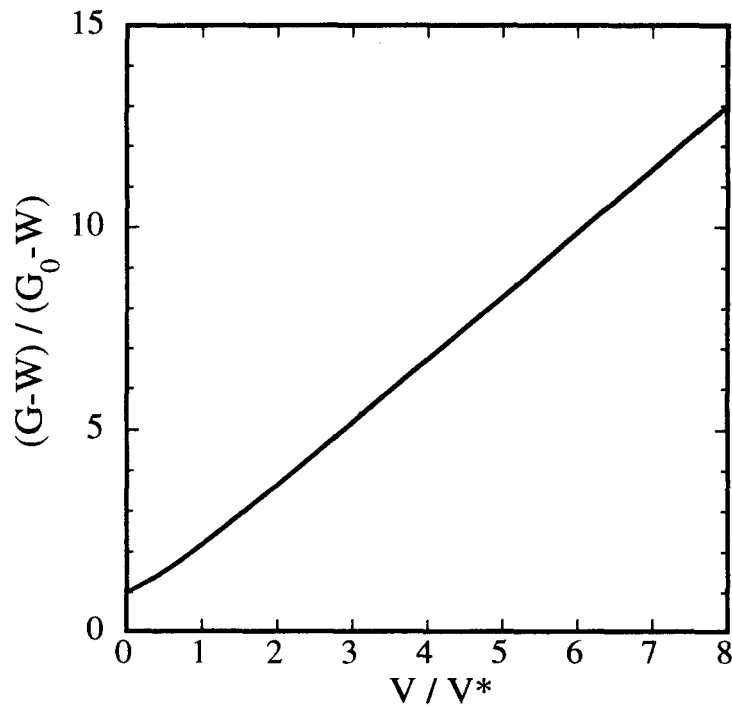


Figure 3. Plot of $(G-W)/(G_0-W)$ versus V/V^* .