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### **Research Article**

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## Peeling Reveals Problems of Strength Standards for Brittle Materials

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#### ABSTRACT

Peeling creates a ubiquitous crack used regularly to open plastic packaging. Such peel cracks have been utilised for centuries, especially in separating cellulose fibres, yet these peeling cracks are not usually mentioned in fracture texts [1,2]. In this paper, theories and experiments show that peel cracks have unique properties quite different from the tensile-test cracks generally used to define and standardize strength of materials. Peeling is the easiest fracture test at the lowest force with the simplest theory describing constant speed cracks that are readily observed. It differs markedly from the catastrophic tensile cracks seen in many brittle materials such as concrete and glass, that accelerate rapidly at high force. The problem is that strength in peeling is not consistent with strength in tension. There is a large size effect in peeling where strength drops for larger sample size. Therefore, standards need to be modified. This paper describes peeling observations in several crack test geometries, fitting the energy theory of thermodynamic crack equilibrium, contrary to the stress/strength criteria that dominate existing brittle-material standards, which need correction.

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#### Introduction

One might wonder why the best-selling book on Fracture Mechanics does not mention peeling in its 611 pages [2]. Yet a peel crack is potentially dangerous because a solid product is weakest in this failure mode, and fracturing suddenly can cause problems. Meanwhile, many materials scientists use peeling crack tests to further a multiplicity of applications, particularly adhesive tapes, which feed a global market approaching \$80billion [3].

Peel cracks have been utilised practically for millenia, especially useful in separating strong cellulose fibres from plant-stems like hemp 10,000 years ago, now exploited in sustainable composite panels for vehicles [4]. All of us have direct experience of peel cracking because we pull apart plastic packaging almost daily. The reason for general engineering ignorance of peeling is that the peel test has been neglected by crack experts, even though it is the easiest crack test to perform, with the simplest theory, at the lowest force, often on transparent tapes where the crack can clearly be seen through the material and observed moving steadily under constant load, surprisingly different from the standard catastrophic tensile crack originally analysed by Griffith in 1920 [5].

The main issue has been that most fracture mechanics scientists are trained in the historic experiments and theories of Griffith, Irwin/Orowan, and others working mainly on tensile stress tests of metals and concrete that crack explosively at high force, causing windows to shatter, ships to sink, bridges to collapse, and aircraft to crash [6-7]. Such tensile experiments have never fitted the Griffith

equilibrium theory of rupture because the measured fracture energies are several orders of magnitude higher than atomic bond energies. Griffith did state that 'work must be done against the cohesive forces of the molecules' in his original paper, but recent textbooks have bypassed atoms, instead emphasising stress analysis rather than the necessary energy balance for dynamic thermal equilibrium of intermolecular bonding [8].

Peeling is more obviously dependent on atomic attractions because you can often see the peel crack moving through the adhered interface without causing any damage to the very smooth, elastic polymer surfaces, which may then heal together again, as in resealable food packs, suggesting a reversible process near thermodynamic equilibrium. Such peel is more often used for cling-film, adhesive tapes, wound patches, and testing of laminates and carbon fibre composites where the components stick together by van der Waals attractive forces. More recently, peel studies have focussed on reversible adhesion where a gecko walks across ceilings by peeling, then by healing its nano-scale footpads back onto the solid surfaces [8]. Here, we view peeling as a direct measure of van der Waals attractive energy between molecules.

Metals displaying plastic flow and concrete crumbling around a crack do not heal like smooth soft polymers and therefore cannot normally be treated with an equilibrium theory of fracture. So, Fracture Mechanics has drifted away from the fundamental ideas of atoms and equilibrium, which are not emphasised in the Wikipedia page, nor in recent books [9-11]. By contrast, this paper views peel cracks as solid state chemical reactions between rubber and substrate molecules, where the dynamic molecular interactions are vital in both peeling and healing. The intermolecular attractive energies then fit the mechanics of thermodynamic equilibrium for

very slow cracks. Faster cracks cause energy dissipation away from equilibrium, addressed in section 3.

The main crack defined in fracture books is tensile fracture, for example of a steel ship breaking due to an explosive crack event [1-2]. The difficulty is that steel ships crack well away from the Griffith theory of equilibrium, and so could never fit the original Griffith mathematics. Contrasting with this lack of agreement between tensile crack results and Griffith equilibrium theory, some peel cracks using adhering soft solids like rubber have been shown to attain true thermodynamic equilibrium and have proved that crack healing can occur and that the measured values of surface energy can match the theoretical energy balance predictions, giving us confidence that peeling can be a truly reversible thermodynamic process [12-14]. Of course, peeling resistance in engineering applications often needs to be increased, for example in durable automotive products, well above the low peel toughness values around 0.1Jm<sup>-2</sup> near equilibrium. Several crack-stopping mechanisms have been found to achieve such toughening requirements for engineering adhesives that need 1000Jm<sup>-2</sup> and more. But then the surfaces are so damaged by the dissipation mechanisms that reversible healing is impossible. The problem of applying an equilibrium theory to experiments that display high fracture energies by dissipating huge amounts of energy is assessed in section 7.

This paper describes three different peel configurations both experimentally and in theory, all differing from the Griffith tensile crack solution. The objective is to headline misnomers like 'peel strength' described in standards, that make smaller test samples seem stronger. Other common errors are 'strain energy release rate' and 'critical stress intensity factor' which do not apply to peel cracks. The first peel geometry considered, in which a shrunk rubber strip peels spontaneously from glass, produces a Griffith type equation but its meaning is different from that found in his 1920 energy analysis [5]. The second test, a 90-degree peeling crack, is totally different from the Griffith/Irwin crack and leads to contrary conclusions because stress has no bearing on the calculation of crack equilibrium force. The third peel test, low angle peeling, demands three terms in the energy equation of state, rather than the two terms always used in Fracture Mechanics since the Griffith original contribution. These three terms lead to a quadratic equation in peel force rather than the square root result first identified by Griffith. This quadratic equation fits the experimental results. Finally, it is essential to understand nonequilibrium peel cracks at higher force and faster velocities, well away from thermodynamic equilibrium conditions. The usual approximation of adding plastic work to thermodynamic surface energy is shown to be unsatisfactory, because three dissipation terms are needed for peel cracks, not just one.

#### **Historical Background**

Fracture Mechanics has a much-documented background based in Griffith's experimental results on fracture of pressurised precracked glass tubes [1,2,11]. He showed failure stress falling for longer pre-cracks, as failure stress  $\sigma$  changed with crack length c in the (corrected) Griffith equation

$$\sigma = \{ EW/\pi c \}^{1/2}$$
 (1)

where  $\sigma$  is the uniform far-field stress assumed in the infinite stretched thin elastic sheet, W is the thermodynamic surface work of adhesion ( $2\gamma$  in the original paper) and c is half the central crack length.

One issue with Griffith's analysis was that he (followed by Irwin and others) only considered two energy contributions, surface energy and potential energy, which turns out to be misleading because two different 'potential energy' terms can be identified: elastic strain energy in the deformed sample; and potential energy in the applied forces [8]. Because the Griffith analysis resolved into a balance between crack surface energy and elastic strain energy at equilibrium, Fracture Mechanics has focussed almost entirely on such cracks, while omitting the potential energy in other applied forces. In addition, Irwin/Orowan introduced a large plastic energy dissipation term, added to the solid surface energy W in equation 1, to match experimental toughness results on metals, up to ten thousand times higher than expected from atomic cohesive energies [8]. The third issue is that Griffith made several errors which made him think his experiments fitted the theory, but he had an order of magnitude error which later came to light. These subsequent results showed that the experiment was not at equilibrium as he had first postulated. In other words, Griffith theory failed to describe his practical measurements accurately. Fourthly, Griffith believed the intrinsic strength concept (ie ultimate strength) formulated by Galileo Galilei who suggested that there was a maximum stress that a material's atoms could withstand, a 'stress criterion' idea which is inconsistent with the 'energy criterion' of fracture that Griffith was proposing [15]. Finally, Griffith was very concerned about small defects, like scratches on the surface, that he believed caused stress concentrations that reduced strength (breaking force/ area) of a solid sample and led Griffith to propose the concept of 'flaw statistics' to account for strength results on fine glass fibres that did not fit equation 1. Here we measure one large crack only, thus eliminating any 'flaw statistics' argument.

Peel cracks have a history that is parallel to Griffith cracks, but the peel crack has not converged with the tensile crack for the reasons outlined below. The first peeling scientist was Obreimoff in 1930 who convincingly explained mica flaking using a bentbeam energy analysis [16]. Obreimoff had peeled thin uniform flakes of mica from a block, then wedged them open and studied the interference fringes in the narrow gap between the crack faces. This was a most remarkable paper because the peeled mica was atomically smooth and jumped back into contact to heal almost perfectly. He had attained equilibrium in fracture measurements for the first time. Moreover, Obreimoff calculated the energy balance using simple bent-beam theory, so he knew both the force applied and the stored strain energy. He matched the energy input to the new crack surface energy created, showing the fracture energy to peel was 1.2 Jm<sup>-2</sup> and that to heal was 0.8 Jm<sup>-2</sup>. The equilibrium value was clearly between these values, about the 1 Jm<sup>-2</sup> energy expected. Although Obreimoff did not mention Griffith, he had used the Griffith energy conservation principle while neglecting crack tip energy, yet still got the right answer by considering only the energy terms distant from the crack tip. In other words, he showed that bending and peeling were much simpler and easier than Griffith tension which required a complex stress analysis around the crack in the test sample. He also found the experimental effects of dwell-time, crack speed and adhesive hysteresis and showed that vacuum increased the fracture surface energy generating electrical sparks, observations never made by Griffith. There was also a size effect, seen but not explained satisfactorily by Griffith: Smaller samples appeared stronger. The Obreimoff energy theory predicted this size effect because he calculated volume strain energy being converted into surface energy, such that smaller volumes contained less energy to feed into the new crack surface, thus requiring higher cracking stress. To summarise, peeling is superior to Griffith and related tensile crack mechanics in several ways:

- Peeling is a crack mechanism where molecules are often reversibly pulled apart against van der Waals attractions, whereas Griffith cracks in glass were not reversible.
- Equilibrium can be demonstrated in peeling; healing occurs; there is no plastic flow.
- Theory of crack equilibrium fits experiments on very slow peel cracks, but never in tensile cracks.
- Peeling crack speed increases steadily with peel force whereas tensile cracks go bang.
- Fracture surface energy measured is correct, not 10<sup>4</sup> times too high as in tensile fracture of steel.
- Simple stress analysis suffices to calculate the energy balance, not possible in tensile cracking.
- Crack tip stresses can be ignored completely in peel but not in tension.
- Dwell-time, crack-speed drag, hysteresis, vacuum and electrical effects are revealed.
- Size effect is clearly defined in peel, where smaller samples need higher stress to drive cracks.

Then, in 1944 Rivlin rationally described how sticky surgical tapes could be rolled onto a polished stainless-steel plate and peeled off at steady velocity by a hanging weight [17]. He first used the simple energy conservation principle shown below in section 5, eliminating elastic strain energy, in which the potential energy delivered by the hanging weight was injected fully into the surface energy of the peeled area. This idea broke away from the fracture mechanics convention that elastic energy is the driving force for all cracks, as in Griffith analysis. Hence, we can delete the misnomer 'energy release rate' which does not exist is simple peel but dominates Fracture Mechanics [1.2,11].

It was not until 1971 that the most significant breakthrough came in the theoretical understanding of peeling and healing smooth spheres from each other or from glass and PMMA surfaces [13]. Optically smooth rubber was used first by Alan Roberts, showing equilibrium with a fracture surface energy of 0.071 Jm<sup>-2</sup> measured at very low crack speed. Immersing in water dropped the fracture surface energy to 0.0068 Jm<sup>-2</sup> which fitted results on water contact angle using Young's equation.Solidified gelatine solutions also showed similar results, with fracture surface energies near 0.105 Jm<sup>-2</sup> as anticipated for van der Waals attractions. Dwell-time, crack speed drag and hysteresis effects were also found.But the biggest shock was the JKR solution to the conservation of energy equation which defined the equilibrium force F to separate two equal spheres [14] as

$$F = 3\pi WD/8$$
 (2)

where W is the thermodynamic work of adhesion and D is each sphere diameter. Unlike the Griffith equation 1, there is no strength here because the fracture force does not depend on area, nor on elastic modulus, and the force is a direct measure of surface energy W. In addition, there is a size effect because small spheres stick much more strongly than large spheres, as proved experimentally. And the mathematical solution gave a quadratic because of three energy terms rather that the simple square root of Griffith, Irwin and followers. 50 years later, this advance still conflicts with conventional Fracture Mechanics. Recent international conferences have been organised and published on this JKR topic to explain its usefulness and to distinguish it from Griffith/Irwin [18]. Sphere testing worked well but smooth rubber strips proved even easier. In 1971 experiment and energy-balance theory showed that peeling smooth strips from smooth Poly Methyl Meth Acrylate (PMMA) or glass surfaces confirmed the sphere peel/heal results, with well-defined equilibrium and the simplest possible solution to the energy conservation equation [13]. Following that breakthrough, this paper brings together different peel test geometries to describe three different ways of peel testing smooth elastic rubber strips from optically smooth PMMA and glass substrates, to give three separate peel crack solutions to the energy conservation principle, all different from equations 1 and 2. We first postulate that crack equilibrium is observed near zero crack speed, where molecules are in dynamic thermal equilibrium, peeling/healing balancing as a consequence of van der Waals attractions. As crack speed is increased, in quasi-static experiments, extra peeling force is required to overcome the attraction energy barrier, as described in section 3.

#### **Importance of Crack Speed**

A vital difficulty of traditional fracture mechanics texts is that they do not consider that the force pulling atoms apart must increase as the crack velocity rises [1,2,11]. Typically, when glass under tensile stress breaks, the crack accelerates as the force drops, which is the opposite of the expected behaviour. We need to show that solid atoms separating under controlled conditions must give crack velocity rising with force.

A typical fracture mechanics book describes cracks moving in several different ways at various speeds [2]. There are catastrophic accelerating cracks, slow moving cracks under creep or corrosion conditions, and fatigue cracks that start and stop under fluctuating loads. The connection between the force separating the atoms and the speed of separation is not mentioned.

Peeling polymers showed this connection in 1956 when Derjaguin's group published the results shown in Figure 1 [19]. Cellulose nitrate film was peeled from glass over a wide range of speeds and the force was measured, showing that the fracture surface energy was 0.04 Jm<sup>-2</sup> at very low speeds, near expectation for van der Waals equilibrium energy, but rose by three orders of magnitude as the crack velocity was increased a million times. It was not clear what caused this increasing energy dissipation at higher crack speeds until studies of several polymers peeling from glass showed that this was a general phenomenon that could be explained by the energy barrier at the surfaces that had to be overcome by activated bond-breaking that might be described by an Eyring activated bond-breaking mechanism described in equation 3 [20].

$$F/b = W + Asinh^{-1}BV = 0.04 + 5 sinh^{-1} 0.025V$$
 (3)

where F is the peel force, b the width of the polymer film, W is the thermodynamic work of adhesion, V the crack velocity with A and B as constants that relate to the interfacial van der Waals bonds. This equation follows from the idea that the adhesive interface has two metastable states with an energy barrier which must be overcome by the applied force assisted by Brownian energy kT, according to the Eyring concept of bond breaking. Thus, the work of adhesion W describes the reversible component, while the term A sinh<sup>-1</sup> BV shows the molecular adhesion crack speed dependence on temperature variant constants A and B.



**Figure 1:** The effect of crack speed on smooth polymer peel energy showing dominance of irreversible peel losses at high speed giving 3 orders of magnitude toughening.

Although the scatter was large, the experimental results approximately fitted equation 3. The polymer film material was elastic and showed no evidence of plastic flow nor surface damage. Healing was possible when the film was brought back into contact with the glass. This reversible thermodynamic W concept in equation 3 together with the energy lost overcoming the molecular bond energy barrier is very different from the Irwin/Orowan idea that the fracture energy is independent of intermolecular forces and depends only on plastic deformation loss.

While metals show very complex plastic behaviours, polymers are often much simpler because they can be almost perfectly elastic, giving predictable and readily understood features. Typical elastomers like natural or synthetic rubber (eg EPA, silastomer or cling-film) can be made optically smooth by crosslinking and cooling against a smooth substrate, and may then be peeled and healed multiple times to give reliable results, while observing the crack accurately through the transparent materials. As we all know from peeling apart plastic packaging, there are several obvious properties:

- 1. The healing process takes some time, usually hours, to reach its ultimate value; this is the dwell-time effect that can be explained by escape of contaminating molecules from the interface.
- 2. The faster you peel, the higher the force required; this is the crack drag effect that was mentioned by Rivlin in 1944, leading to a characteristic peel energy versus crack speed curve, as shown in Figures 1, 3,5, and 7 [20]. Such characteristic curves do not exist in Fracture Mechanics texts where plastic flow is the dominant phenomenon [1,2,11].

The preliminary conclusion is that reversible thermodynamic equilibrium is the prime concern, followed by the lossy effects of cracks speed, then later by irreversible relaxations like plastic flow or viscoelastic droop. Now we consider both the main features in several different peeling configurations.

#### Peel Crack Driven by Stored Elastic Strain Energy

The peel crack configuration shown in Figure 2 allows a prestrained rubber strip to peel spontaneously away from its glass substrate without applying any external force, so is similar to the 'fixed grips' situation emphasised by Griffith, Irwin and followers, in which two energy terms, elastic and surface energy, are exchanged, without any applied force requiring an extra potential energy term. Thus, it is fair in this peculiar geometry to discuss strain energy being converted exactly into surface energy at equilibrium, or into surface energy and dissipated heat away from equilibrium at high crack speeds. Still, the Irwin concept of 'energy release rate' needs to be deleted because it is not time dependent, nor is it released: It is converted fully into surface energy of the molecules at equilibrium, with further heat production at high crack speeds.



**Figure 2:** Experiment to demonstrate spontaneous peeling of smooth rubber film, width b, thickness d, with residual tensile strain  $\varepsilon = e/z$ . a) free smooth rubber film; b) stretched film with displacement e to strain  $\varepsilon$ ; c) adhered stretched film to smooth glass substrate with both ends unstressed (grey colour); d) spontaneous peel cracking of film without any applied force.

The energy balance theory fitting this peel crack behaviour was first derived in 1973 and shown to depend on two energy terms only, a surface energy term and an elastic strain energy term, akin to the Griffith theory of 1920 [21,8]. The surface energy is

 $U_c = Wbc$ 

where c is the crack length and b the strip width (Figure 2) while W is the thermodynamic work of adhesion.

The elastic strain energy term is

 $U_e = -\sigma^2 bdc/2E + crack tip energy field$ 

where E is the Young's modulus of the rubber and  $\sigma$  is F/bd with F the stretching force applied as the strip of thickness d was healed onto the glass surface.

This gives the total energy

 $U_{T} = Wbc - \sigma^{2} bdc/2E + crack tip energy field$ 

So, application of the Griffith energy conservation principle gives  $dU_T/dc = 0 = Wb - \sigma^2 bd/2E$ 

because the crack tip field disappears as it is constant for a long crack, so that the equilibrium peel crack equation is

$$\sigma = \{2EW/d\}^{1/2}$$
(4)

which has the same dimensions as Griffith equation 1 but is totally different in its import because the stress is stretching the film longitudinally, not pulling it at right angles from the glass substrate. So, a peel cracking strength cannot be defined because the stress  $\sigma$  is not linked to the area of contact between film and surface. This is the main reason why the definition of 'peel strength' in several ASTM standards is inadequate as discussed in section 9.

Also, the peel crack travels in the same direction as the applied tensile stress, rather than perpendicular to it like Griffith. In addition, the crack criterion does not depend on crack length. The crack therefore travels at constant speed, unlike the accelerating Griffith crack. Moreover, the stress field around the crack tip is irrelevant and there is no need for a 'stress intensity factor' which had been defined to fit the energy balance result for the Griffith crack geometry [1,2,11]. Clearly, this peel crack is driven only by the tensile strain energy, has nothing to do with shear or peel strength, and should be called tensile delamination, as defined for adhesive lap joints [22].

In retrospect, it is obvious that a two-term energy equation based on W and  $\sigma^2/E$  must lead to solutions like equations 1 and 3, and some different examples exist but several do not have a 'critical stress intensity factor' and do not depend on  $c^{-1/2}$  [23]. The conclusion must be that the Griffith equation 1 is a particular, unique and special solution of the energy conservation equation described in Fracture Mechanics text books that ignore the many other dimensionally similar but practically different particular solutions, seen in peel, lap joint failure, JKR, bending cracks etc. The most striking idea emerging from equation 4 is that brittle strength  $\sigma$ , envisaged in the Griffith equation 1, does not apply to peel cracking. Moreover, such a strength would have to increase as film thickness d falls. Thinner films seem to stick stronger. This is the size effect emphasised in [23] because small brittle samples usually exhibit higher strength, but ignored in all other Fracture Mechanics books except Bazant et al [1, 2, 11, 23].

Experimentally, a transparent smooth rubber film was made by hot-pressing a natural rubber compound onto a glass sheet to form a film 0.2mm thick, and allowing sulfur crosslinking to solidify the film, which was then cooled, released and cut into 10mm wide strips, which were healed into contact with smooth glass substrates. As shown in Figure 2a, a strip of length z, width b and thickness d, was selected and first pressed into contact with the glass surface, making sure to avoid trapped air bubbles and any residual strain. After 2 hours of contact to allow any gas to diffuse out, the adhesion reached a steady value and was measured in the 90° peel test (Figure 4) by attaching a weight to the film and measuring the resulting crack speed by observing the peel crack through the glass. The bold curves in Figure 3 show the results of these experiments, showing that peel cracking can be defined by a characteristic peel-energy versus log peel-speed curve, in which the low-speed peel-energy is near equilibrium, while high speed cracking requires higher peelenergy away from equilibrium.At low weight/strip width, below 0.3g/cm (using equation 4:F=Wb), the crack did not propagate, and at lower weight, about 0.1g/cm, healing could be seen, indicating that equilibrium conditions were attained with thermodynamic work of adhesion near W= 0.2 Jm<sup>-2</sup>, as expected from van der Waals attractions. As bigger peeling energy was applied, the crack increased in speed, indicating that energy was now being dissipated into heat, thermal energy ten or hundred times bigger than W. The peel energy F/b was then called R to distinguish it from true equilibrium energy W.The objective of the experiments was to test equation 4 by carrying out the steps shown in Figure 2b, c and d. A rubber strip was stretched to extension e, strain  $\varepsilon$  and stress  $\sigma$ before healing to the glass as before.

To obtain the results of Figure 3, force was first applied to the strip to extend it by strain  $\varepsilon$ , then the prestrained strip was rolled into contact, pressing the ends to give unstressed regions that formed interface dislocations, thus stopping premature peeling. Figure 2b shows the uniform pre-stressing and Figure 2c shows

the stretched film after healing into perfect contact with the glass substrate. Both ends of the strip are coloured grey because these parts were unstressed and formed adhesive dislocations to prevent instantaneous fracture. There was a dwell-time effect, so the films were left in contact with the glass for 2 hours to attain a constant adhesion energy. Temperature was also important because the characteristic peel energy versus crack speed curve moved to higher speed as temperature increased. All experiments were at 20°C.

A test was carried out by defining the prestress in terms of prestrain, then tugging at one end of the film to push a starter peel crack past the dislocation, then observing the crack through the transparent glass to measure crack speed.Below prestrain  $\varepsilon$ =0.06, the peel crack did not propagate spontaneously. The conclusion was that the elastic strain energy was less than the surface energy required by the crack opening. At low prestrain  $\varepsilon$ =0.06, the crack only advanced at very low speed, less than 10 µms<sup>-1</sup>, and so was judged to be near equilibrium. This is the left-hand point in Figure 3a. This result fitted equation 3, proving that peeling does not always need an applied force but can be driven by stored elastic energy.

With higher prestrain  $\varepsilon$ =0.13, the film peeled off spontaneously, but the peel crack moved much faster, almost 1mms<sup>-1</sup>, and thus could not be at equilibrium so equation 3 could not apply. Clearly, the stored strain energy was far higher than the required crack surface energy and heat was being dissipated. Therefore equation 4 was modified to

$$\sigma = \{2\mathsf{E}\mathsf{R}/\mathsf{d}\}^{1/2} \tag{5}$$

where R was the measured fracture energy, not the equilibrium work of adhesion W. Using equation 5 to calculate R from the known residual stress  $\sigma$  showed that the peel energy was equivalent to F/b of 2.2g/cm, nearly fitting the measured F/b versus crack speed line. Higher prestrain  $\epsilon$ =0.22 made the film peel spontaneously still faster at 3780µms<sup>-1</sup>, somewhat less than 6000 µms<sup>-1</sup> calculated from the equivalent 6.3g/cm F/b.

Figure 3b shows results from testing three thickness of rubber film, 0.2mm, 0.4mm and 0.6mm, all carried out at prestrain  $\varepsilon$ =0.13. As expected, the larger thickness contained more elastic energy and therefore peeled off faster to dissipate more heat. The three experimental points calculated from equation 4 fitted the calibration curve reasonably well, showing that the prestrain was equivalent to a calibration peel energy F/b by equation 5.



**Figure 3:** Results for spontaneous peeling of prestressed rubber strip from glass. The bold lines are characteristic peel-energy (W=F/b) versus crack speed V curves showing the peel results at zero prestrain, attaining equilibrium about  $1\mu$ ms<sup>-1</sup> crack speed; a) Experimental points show results for spontaneous peeling of 0.2mm thick films for three prestrain values; b) Points show results for spontaneous peeling at prestrain 0.13 for three different film thicknesses.

To summarise these results, elastic strain energy can drive a peel crack without application of any external force. Small prestrains do not cause peeling. Increasing the prestrain did produce equilibrium cracking, while larger prestrains drove the crack faster to dissipate heat, away from equilibrium but fitting the peel calibration curve relating peel-energy to log(crack-speed).Peel strength is a misnomer because the prestrain required for peeling does not rise with contact area. However, this prestrain did rise for lower film thickness, giving a size effect: Thin films stick better. The main advance is that equilibrium equation 4 works at very low speed while equation 3 gives fair agreement if the crack speed, ie the rate of molecular separation, is controlled.This concept differs from Griffith who did not mention, nor measure, crack speed, nor Irwin who merely added a large plastic term to W to give R.

Peel Crack Driven by Potential Energy in the Applied Force Our second peel configuration is shown in Figure 4, which shows a smooth silicone rubber strip healed onto a PMMA substrate and then peeled by an attached weight. This is the standard peel calibration test defining the characteristic peel-energy versus log (crack-speed). The peel force F was applied on the strip width b at right angles to the substrate to bend the film without significant elastic stretching. A hanging weight was used. The peel crack was viewed through the PMMA and seen to move at constant speed under steady load. At low force, there was no crack propagation, but crack healing was observed instead when the weight per cm width was reduced below 0.12 gcm<sup>-1</sup> (Figure 5). Between 0.12 and 0.3 gcm<sup>-1</sup> the peel crack movement could not be detected. This was adhesion hysteresis. Somewhere in this zero crack speed regime was the equilibrium work of adhesion W. This was the equilibrium point where peeling and healing were in balance. Increasing the force/cm above 0.3 gcm<sup>-1</sup> caused propagation at higher constant speed, indicating that energy was being dissipated as the surfaces were separated, producing heat away from thermodynamic equilibrium. There are three key questions: First, is energy dissipation occurring at the interface where atoms are being separated? Second, is some of the dissipation at the interface and some within the material bulk? Third, since the system is now away from equilibrium, can the thermodynamic equations work?



**Figure 4:** Smooth rubber strip peeling perpendicularly from PMMA under a weight F with crack length c.

Three energy terms are needed. The surface energy is  $U_s =$  Wbc where b is the strip width and c is crack length, potential energy is

 $U_n = -Fc$ ,

and elastic energy is

 $U_e$  = constant energy in the bent film and in the crack tip field which does not change at constant force and steady crack speed. Applying the equilibrium energy conservation criterion, the constant elastic energy  $U_e$  disappears

$$d/dc [Wbc -Fc + U_{e}] = 0$$

$$\mathbf{F} = \mathbf{W}\mathbf{b} \tag{6}$$

which is the simplest possible crack equilibrium equation, totally different from equation 1 because elastic energy has no influence on this peel crack. The main point in this analysis is that most of the elastic energy is in the bent film, which is so thick that it does not extend significantly, and this bend energy remains constant as the crack moves, so disappears on differentiation to define the equilibrium, because total energy must be conserved. The crack-tip strain energy also has no effect on the crack, since it also remains constant as the crack moves at equilibrium, so disappears on differentiation. The conclusion is that the crack is driven only by potential energy in the force F. The crack is not influenced by strain energy and so has no connection with stress, with Griffith equation 1 nor Irwin's  $K_{1e}$ .

This peel crack theory is totally different from the standard fracture mechanics tensile crack based on Griffith geometry.Whereas the equilibrium Griffith crack equation 1 defines stress  $\sigma$  as the measure of  $\{W\}^{1/2}$ , the peel crack uses the applied line-force F/b as a direct measure of W. This peel line-force does not change with crack length c. F is independent of contact area so stress is not of interest, while elastic modulus is irrelevant.Because this simple peel crack does not depend on c<sup>-1/2</sup> (which applies in Griffith equation 1) the concept of 'critical stress intensity factor' K<sub>1c</sub> is inapplicable.Instead, the peel line-force is a direct measure of fracture surface energy and completely independent of stress anywhere in the sample. Also, there is no 'strain energy release rate' because strain energy remains constant throughout at equilibrium. ASTM standards mention 'peel strength' but that needs changing (see section 9) because peel force is independent of contact area. However, if you try to pinpoint an area that is acted upon by the peel force, there is a size effect because peel force remains the same experimentally as film thickness is reduced.

Figure 5 shows typical peel force versus crack speed results showing that equilibrium can be defined at very low speeds around 10nms<sup>-1</sup>. Crack drag was most significant as peel force rose rapidly with crack velocity, showing that there was interface heating proportional to rate of molecular separation. There was also crack hysteresis, that is a gap in force between peel and heal forces. However low the crack speed, it was almost impossible to hit true equilibrium, even for very elastic rubber. Slight dissipation caused crack stopping before equilibrium could be established (see Figure 7). Making the rubber less elastic gave an increase in this hysteresis.



**Figure 5:** The effect of crack speed on smooth silastomer peel energy showing the equilibrium value W and irreversible energy losses at crack speed V. The healing curve proves that thermodynamic equilibrium is achieved, but some adhesion hysteresis prevented observation of the true equilibrium W. Hysteresis increased as the rubber was made less elastic.

#### Peel Crack Driven by Both Potential and Elastic Energy

Rivlin did not mention Griffith in his short paper on 'The effective work of adhesion' but was correct in showing that simple peel converts potential energy in the applied force into surface energy of the crack [17]. Later in 1971, a concept emerged that was entirely different from the Griffith/Irwin idea that elastic energy was the main term driving the crack, always equal to half the potential energy in the applied force [13]. Instead, a thermodynamic theory was defined with total energy  $U_T$  given by the sum of three energy terms, surface energy, potential energy of the applied force and elastic strain energy in the deformed rubber

$$U_{\rm T} = U_{\rm s} + U_{\rm p} + U_{\rm e}$$
 (7)

In which  $U_p$  was not twice  $U_e$ . This concept of three independent energy terms has been successful describing JKR and other cracking geometries, very much differing from the Griffith equation 1 of 1921 [5]. Griffith was dominated by elastic strain energy, a particular solution not generally applicable to geometries like peeling or lap joint cracking. Whereas the Griffith crack geometry has never been shown to reach equilibrium, peel and lap cracks have all demonstrated peel/heal equilibrium [8].

Figure 6 shows how three independent terms can be found in a certain type of peel test in which the peel angle  $\theta$  is gradually reduced to low levels. Rivlin first used energy conservation to demonstrate that the peel force F must be increased from equation 6 to

$$F = Wb/(1 - \cos\theta)$$

The free rubber peel arm, at low angles, then began to extend elastically under higher force as in Figure 6a and demanded an extra elastic energy term that increased in size as the peel crack propagated, unlike the 90-degree bend energy and crack tip energy that remained constant and consequently disappeared on differentiation. The small elastic extension of the film element length z is e which can be worked out from the equation E=stress/strain=(F/bd)/(e/z)

$$e = Fz/bdE$$

where the Young's elastic modulus is E.Consequently, the force moves further than before to give an extra potential energy

$$U_{p} = -Fe = -F^{2}z/bdE$$

The elastic energy stored in the film is now increased by half this value, increasing the total elastic energy in the film to

 $U_e = F^2 z/2bdE + bending energy + crack tip energy$ 

Therefore, the total energy in the system is  $U_p + U_e + U_s$  $U_T = -F (1-\cos \theta)z - F^2z/bdE + F^2z/2bdE + bending energy/crack$ tip energy + zbW

= -F  $(1-\cos \theta)z$  - F<sup>2</sup>z/2bdE + bending energy/crack-tip energy + zbW

Applying the equilibrium condition with constant bending/cracktip energy

$$dUT/dz = 0$$
 = -F (1-cos  $\theta$ ) - F<sup>2</sup>/2bdE +bW

Thus, the equilibrium of this crack is given by a quadratic equation in F/b first found in 1971-1975 [13, 24].

$$(F/b)^2/2dE + (F/b)(1-\cos\theta) - W = 0$$
 (8)

a quadratic reminiscent of those found in JKR cracks [14] and compression splitting cracks [25].

At constant force F, the peeling occurs at constant speed as in sections 4 and 5, but the interesting feature is that the strain energy release rate is negative, as in many fracture test experiments like Obreimoff and Benbow/Roesler double cantilever bend (DCB) tests [16,26]. But more potential energy is injected, making the joint weaker as this extra energy is converted to new crack surface. Irwin and his followers never mentioned negative strain energy release rate, which throws a new light on fracture mechanics terminology that should be deleted.



**Figure 6:** a) Elastic extension of the film (magnified below) at force F changes the energy balance; b) experimental results compared with equation 8, showing how the force levels off at low peel angle where elastic energy dominates. The thermodynamic theory fits the experimental results, though far from true equilibrium, with controlled peel crack speed 0.08mms<sup>-1</sup>.

Equation 8 was tested experimentally using EPR transparent elastomer Enjay 404, crosslinked with 0.32 % sulphur and 2.7 % dicumyl peroxide against a glass plate at 160°C for 1 h. After this sheet was peeled from the glass when cooled, a smooth rubber surface was revealed that could be re-adhered to the glass to give reproducible adhesion. In the present tests the rubber was readhered for a contact time of 1 hour to ensure that all surface gas molecules had escaped to reach van der Waals adhesion conditions. The rubber thickness d was 0.75 mm and Young's modulus E was 1.21 MPa. Measuring the peel cracks near equilibrium around 0.001 mms<sup>-1</sup> proved to be too slow in practice. Instead, the peel crack speed was maintained at 0.08mms<sup>-1</sup> to ensure that the van der Waals bonds were being broken at the same rate for each peel angle experiment. The experimental points then fitted the equilibrium equation 8 even though it was obvious that the peel energy was a factor 20 higher than the known thermodynamic work of adhesion W value around 0.07Jm<sup>-2</sup>. The conclusion was that the equilibrium equation for peel cracking could be used well-away from the true equilibrium condition, providing the crack speed was properly controlled. The limit to this finding is not known but the experimental peel energy should then be called R to distinguish it from the thermodynamic value W.

Non-Equilibrium Peel: Crack Slowing and Crack Stopping

In reality, most cracks are studied away from equilibrium, where irreversible losses generate heat energy such that more force has to be provided, and this increase can be many orders of magnitude, taking toughness R (measured fracture energy distinguished from equilibrium work of adhesion W) to high levels up to 10<sup>5</sup> Jm<sup>-2</sup>, so high that interatomic forces cannot account for it. Irwin/Orowan took the lead in suggesting that W rose to R because of plastic work J to be added to W, readily seen around metal crack tips such that Griffith equation 1 fitted the crack results, but with much higher effective surface energy R. There is an element of truth in this concept, but we already saw in section 3 that the situation is much more complex because atoms are being pulled apart. During rubber peeling, no plastic flow was detected and healing was observed. Therefore, the explanation of crack drag for highly elastic peeling was the extra force required to break van der Waals bonds in an activated Eyring rate process which gave equation 3, that was found to fit many results in Figure 1.

Formally, equation 7 must be changed because an additional thermal energy term  $U_t$  is needed to satisfy conservation in a non-equilibrium peel model. Total energy is then

$$U_{T} = U_{s} + U_{p} + U_{e} + U_{t}$$
 (9)

in which  $U_t$  can produce heat by two obvious processes: first the interface heating caused by molecule separation, the crack drag effect of equation 3; second the bulk heating in the material due to inelastic relaxation, which can be viscoelastic for polymers, but plastic for metals. For viscoelastic rubber, this can be expressed in terms of the original unpeeled elastic modulus  $E_o$  and the relaxed elastic modulus  $E_t$  of the peeled rubber after a time t. This is essentially a hysteresis loss as the rubber is cycled from zero stress before peeling, to a high stress as the crack hits, to a low stress after the rubber moves fully through the peel bend. The simple result from many peeling experiments was that this thermal effect could be simply defined by multiplying equation 8 by  $E_o/E_t$  giving

$$F/b = (E_o/E_t)(W + Asinh^{-1}BV)$$
(10)

indicating that relaxation in the bulk material has a strong effect when  $E_o/E_t$  is large. This is only a small effect in the case of highly elastic rubber materials because the modulus drops by just a few per cent as the material moves through the peel bend [27].

However, this easy combination of interface loss and bulk loss was found to be incorrect experimentally because there was a crack slowing effect. Peeling smooth rubber strips from smooth glass or PMMA revealed that crack speed did not remain constant under steady force at the low crack velocities around 0.01µms<sup>-1</sup>, an observation that was due to the slight viscoelastic relaxation of the rubber with time, reducing the bending moment of the force applied at the crack opening [8]. Therefore equation 10 is not correct because it assumes the peel bend shape is constant, such that the bending moment applied by the force is unchanged.But experimentally, a freshly peeled element of rubber is drooping viscoelastically, cutting the mechanical bending moment, to slow the crack speed significantly. The effect is shown in Figure 7a that gives results compared to the theoretical line from equation 3 for a silastomer sample peeling from PMMA under constant force of 0.5g.

$$F/b = W + Asinh^{-1}BV = 0.25 + 0.09 sinh^{-1}V$$
 (11)



**Figure 7:** Crack slowing causing crack hysteresis; a) Peel energy plotted against crack speed showing crack slowing observations below peel crack speed 100µms<sup>-1</sup>, giving hysteresis; b) Typical crack slowing curve fitting the equation that includes a drooping term

Initially the peel crack speed was  $10\mu$ ms<sup>-1</sup>, but this halved within one minute and continued to fall over 30 minutes as shown in Figure 7b. At peel speeds above 100  $\mu$ ms<sup>-1</sup>, the crack speed was constant because the rubber was moving too fast through the peel bend to droop enough for slowing the crack. Figure 7a thus illuminates a cause of peel hysteresis. Low speed cracks slow and stop so the measured adhesion energy seems four times higher than equilibrium.

The mathematical description of this crack slowing effect depends on the measured rate of change of the rubber elastic modulus E with time t after load was applied

$$dE/dt = -CE_{o}/t \tag{11}$$

where C is the relaxation constant and E is the Young's modulus at time zero. By considering the energy balance of a short length x of the peeling film suddenly loaded as the crack moves, it was shown [20] that an extra term F(C/t)(x/(dx/dt)) must be added to equation 10 to describe the influence of viscoelastic drooping on the crack energy conservation

$$F/b = (E_o/E_t) \{W + Asinh^{-1}BV - F(C/t)(x/(dx/dt))\}$$
 (12)

where x is a short length of peeling rubber which moves into the crack zone. This new term in equation 12 is fascinating because it grows as the crack slows, leading to a rapid hyperbolic drop in velocity, fitting the results shown in Figure 7b. One solution to this equation is t(dx/dt) = constant which predicts the observed hyperbolic slowing observed. In summary, the traditional Irwin/ Orowan addition of a single large constant R to increase W as an explanation of the huge fracture energy hysteresis seen in metals, is an oversimplification because equation 12 shows that several additional ideas are needed to explain the peel crack results. The bulk effect is given by the factor  $E_{a}/E_{t}$ . The interface mechanism Asinh<sup>-1</sup>BV is the atoms breaking apart across an activated energy barrier. The third unexpected term is the change of the bending moment with time as the viscoelastic film droops under constant force F, to give crack slowing with time. Fracture mechanics has failed to recognise these crack processes away from equilibrium. It has also failed to recognise the equilibrium crack-stopping effect when a peel crack hits a high modulus interface, moving from  $E_1$  to  $E_2$  where W is amplified to  $WE_2/E_1$  first demonstrated in a peeling crack, an effect that can explain the factor 200 toughening observed in a carbon fibre reinforced plastic composite [27]. Sticking brittle glass fibres together with brittle polymer matrix can increase fracture toughness because of modulus mismatch [28].

#### **Standards for Peeling Need Attention**

Equation 8 describing the elastic term in peeling gives a neat result as the peel angle approaches zero (Figure 8a), such that the elastic energy term feeds directly into the surface energy.

$$(F/b)^2/2dE-W=0$$
  
 $F/b = \{2WEd\}^{1/2}$  (13)

The peeling film then looks like the lap joint shown in Figure 8b



**Figure 8:** a) low angle peeling; b) peel at zero peel angle is lap test; c) When healing occurs, an interface dislocation is formed.

This equation was experimentally verified to show that this socalled lap shear joint is essentially a low angle peel test where shear is not relevant. Typical standards for lap shear joints state that the failure force is proportional to the overlap area, whereas many tests have shown that the lap fracture force does not increase linearly as the overlap is extended [22,29,30]. The calculation of lap shear strength using the stress criterion is therefore not valid.

Instead, equation 13 shows clearly that the energy criterion for cracking is correct and has the same units as equation 4 for shrinkage delamination. The failure force depends on thickness WEd<sup>-1/2</sup> which gives a large size effect, with thinner films giving apparently stronger lap adhesion. Consequently, 23 lap-joint ASTM standards need to be modified to avoid large errors, as described in [22].

The key difference between a true shearing process and low angle peeling is illustrated in Figure 8c, where the crack propagates for a certain distance, then the new crack surfaces jump back into contact, with crack surfaces displaced to form an interfacial dislocation [8]. Shearing is not taking place, yet the rubber is moving along the substrate by crack opening, displacement and healing, reminiscent of Schallamach waves first observed during rubber sliding in 1971 [31]. The myth of 'shear' in such complex fracture processes has been discussed since 1998 [32].

Applying similar logic to peel force and equation 6, the concept of peel strength also requires attention mentioned in a recent paper [33]. The main problem is that 'peel strength' is defined in many standards as F/b (peel force divided by strip width) which is not stress but energy per unit of fracture area. Since the ancient Galilean concept of strength is based on the stress criterion of failure, that fracture occurs at a critical maximum stress, it is more rational to define peel strength as the peel force divided by an area, for example F/bd (peel force divided by film width times thickness). We should therefore change these faulty standards and define F/b as peel energy or peel fracture energy, as throughout this paper. It could also be called peel toughness because that is the energy required to separate unit area of adhering contact. Table 1 lists a range of peel standards that need changing to make them consistent with experimental observation and the peel energy units Jm<sup>-2</sup>.

Table 1 List of peel standards that require corrections to remove errors.

ASTM-D1876 – standard test method for peel resistance of adhesives (t-peel test)

ASTM-D1995 – standard test methods for multi-modal strength testing of autohesives (contact adhesives)

ASTM-D3330 – standard test method for peel adhesion of pressure-sensitive tape

ASTM-D5458 – standard test method for peel cling of stretch wrap film

ASTM-D6252 – standard test method for peel adhesion of pressure-sensitive label stocks at a 90° angle

ASTM-D6862 – standard test method for 90° peel resistance of adhesives

ASTM-D903 – standard test method for peel or stripping strength of adhesive bonds

ISO-11339 – adhesives – T-peel test for flexible-to-flexible bonded assemblies

ISO-8510-1 – adhesives – peel test for a flexible-bonded-to-rigid test specimen assembly – part 1: 90° peel

ISO-8510-2 – adhesives – peel test for a flexible-bonded-to-rigid test specimen assembly – part 2: 180° peel

ASTM D1876 is perhaps the most popular test because it requires only two grips to grab the ends of the T peel test-piece, plus a tensile test machine to measure the force at steady speed peeling. Websites describe peel strength whereas some standards (eg D6862) refer to peel resistance and others (eg D903) define stripping strength, which all should be called peel energy [34]. The force is measured to detach the two adhering films for a standard width but the factor 2 is missing for comparison with a 90-degree peel test, using the energy conservation theory [8]. Other peel tests D1995 and D3330 refer to F/b as peel strength but D5458 calls it cling when measuring peel adhesion of very thin polyethylene sheets whereas D6252 on peel testing sticky labels calls it adherence.

ASTM D1995, mainly used for contact adhesives (autohesives), includes D1876 T peel test but also uses compression shear, cleavage and climbing drum peel. Peel strength is calculated from the force divided by strip width, and needs to be redefined as peel energy, not strength [35]. ASTM D3330 is a peeling test for pressure sensitive adhesive tapes and can be 180-degree or 90-degree peeling [36].

ISO 11339 is a T peel test which again calls F/b the peel strength. To summarise, many existing standards for peel adhesion testing remain confused about strength when peel energy ie energy to break unit area, is the term we seek.

#### Conclusion

Peel cracks are highly relevant to plastic packaging and adhesive tapes that we utilise constantly. They are the simplest cracks yet discovered, both experimentally and theoretically, and therefore can be used to question Fracture Mechanics, which is often based only on the Griffith equation. Peel cracks experimentally differ from Griffith because peel cracks propagate at the lowest possible force, move at constant speed and can be observed easily through the transparent polymer-strip, glass or PMMA substrate, delivering a controlled crack speed that increases with increasing applied force, explained by the activated breaking of van der Waals bonds at higher rate. Such crack drag is universally observed in peeling yet never yet found in metal fracture during tensile tests. Healing is also observed with a dwell-time effect of increasing adhesion as contaminant surface molecules diffuse out from the healed interface. Adhesive hysteresis was seen as precise equilibrium of the crack peel/heal was difficult to produce, even with the most elastic rubber samples.

The theory of peeling cracks is based on Griffith's original energy conservation principle but differs totally in geometry, loading and theoretical result, requiring no complex stress analysis, rather like the Wilhelmy plate argument for liquid surface energy measurement, readily understood in secondary school, giving a similar equation for equilibrium peel force F=Wb where W is thermodynamic work of adhesion and b the width of the peeling film [28]. In addition, there is no energy release rate and no critical stress intensity factor during peel cracking.

Three differing peel tests can be distinguished depending on the geometry and loading method. Each peel test gives a different particular solution, none being equivalent to the Griffith equation on which fracture mechanics was founded. To summarise, there are many particular solutions to the energy conservation equation of equilibrium fracture, not just the Griffith equation. Moreover, 'flaw statistics' first suggested by Griffith to explain how small brittle samples appear stronger is unnecessary because the peel solutions naturally reveal such a size/strength phenomenon.

The first peel test, describing spontaneous peeling of a shrunk rubber film from glass, is the nearest to Griffith/Irwin ideas because strain energy is converted into crack surface energy, giving two terms in the energy conservation equation. However, the final equilibrium equation is different from the Griffith equation, though dimensionally similar. The peel crack travels at constant speed and crack equilibrium does not depend on crack-tip stresses. Also, there is a size effect in which thin films appear to stick stronger, requiring higher shrinkage stress to obtain the peel cracking.

The second peel test described has no elastic energy term. It converts potential energy in the applied force into crack surface energy.Hence

it leads to an equilibrium equation different from Griffith. There is no 'strain energy release rate', no 'critical stress intensity factor' and no effect of crack length nor stress on the peel force. Moreover, there is a size effect because thinner films peel at the same force, whereas stress analysis would predict a smaller force.

The third peel test differs from Griffith's original energy analysis because it has three, not two, independent terms in the energy conservation equation: surface energy, elastic strain energy and potential energy of the applied force. It leads to a quadratic equilibrium equation which differs from Griffith, giving a size effect in certain conditions. For all three peeling tests, the stress distribution around the crack can be ignored completely, yet the theory fits the experiments.

Perhaps the most significant result of studying the three peel crack configurations is that peel strength cannot be defined because the peel force does not depend on contact area, whereas the Griffith equation defines a strength (force/area) that is constant if the flaw size remains the same. Peel force varies with sample size, but not with contact area, so the idea of a reliable brittle peel strength is absurd. There is confusion in standards because the peel result F/b is variously called peel strength, peel resistance, cling, adherence or stripping strength. F/b cannot be strength because it does not have units of stress. ASTM and other peel standards should therefore be changed to define F/b as peel energy or peel fracture surface energy or peel toughness, which are all descriptions of energy required to crack unit area of interface. It is clear that peel cracking tests measure peel fracture surface energy (F/b) Jm<sup>-2</sup> as in Figures 1,3,5,6,7, not strength nor stress.

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