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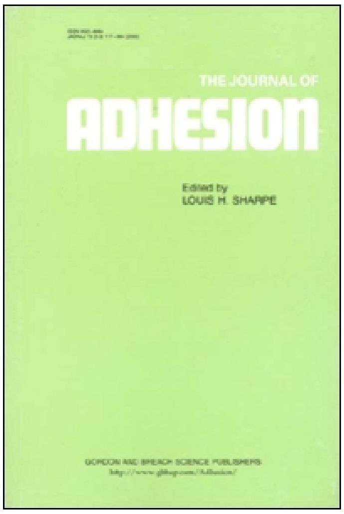

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Consideration of Energy Dissipation for the Strength of Adhesive Joints

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Consideration of Energy Dissipation for the Strength of Adhesive Joints

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For some adhesive joints where the main difference is the degree of contact at the interface, failure occurs not at the interface, but some distance away in the polymer itself. This cohesive mode of failure in the polymer was always found to be the case in our studies of cupric oxide to branched polyethylene interfaces, even where the joint was so weak that the peeled surface seemed clean of the polymer to the naked eye. It was observed that the strength of the joint was associated with the coarseness of the texture of the peeled surface of the polymer. With a differential scanning calorimetry technique we have shown that the coarseness of the surface texture and therefore the strength of the joint, is a direct function of the amount of polymer involved in plastic deformation. The strength criteria for the adhesive joint of this kind is thus the energy of deformation and not the maximum tensile stress that the material can withstand.

INTRODUCTION

Schonhorn and Sharpe[1] have shown that failure of an adhesive joint occurs, in general, not exactly at the interface, but at a small distance from the interface. The failure is thus considered to be a cohesive failure and, in the case of a metal-to-polymer interface, it is the failure within the polymer bulk. The concept of the weak boundary layer was introduced[2,3,4] and fortification of the polymer near the interface through crosslinking[5] or through favorable morphological modifications[6,8] had been shown to result, sometimes significantly, in improving the strength of a joint.

It is the purpose of this paper to present a mechanical model which is based upon an evaluation of stresses in the polymer in a region near the metal oxide–polymer interface. Without any consideration of polymer

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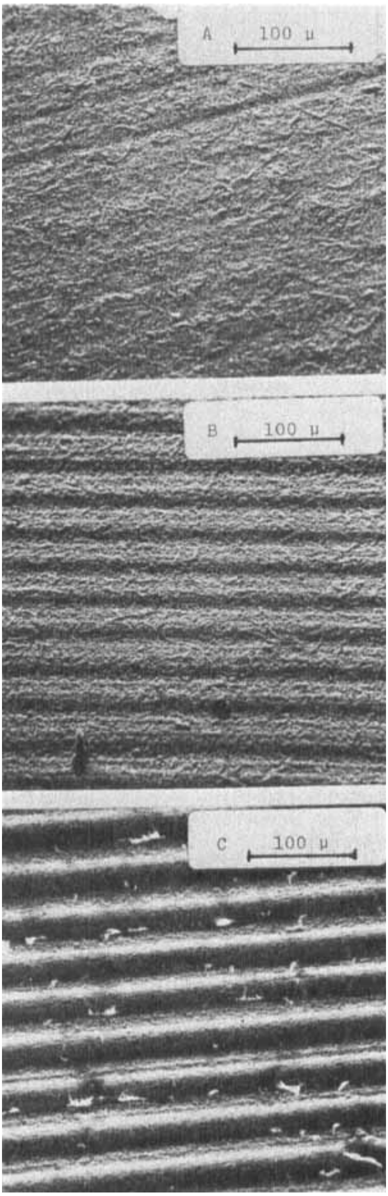

FIGURE 1 Scanning electron micrographs of copper oxide surfaces after polyethylene has been peeled away. (A) Sample 1, hand-buffed prior to processing. (B) Sample 2, adhesive joint was made at inadequately low (95 ~ 105°C) temperature. (C) Sample 3, marginally satisfactory joint.

morphology and taking into account only the amount of polymer which wets the oxide surface, the model defines the thickness of the zone adjacent to the interface where plastic deformation occurs. It was observed that the amount of polymer involved in this drawing process determines the strength of this type of adhesive joint.

EXPERIMENT

In this study we shall consider only adhesive joints between branched polyethylene and cupric oxide (CuO) surfaces, as utilized in the high pressure seal of the ocean cable repeater unit. The metal oxide surfaces were prepared on machined copper plungers by J. Rinaldi using Ebonol C, proprietary material from Enthone, Inc.[7] It is known that if the polyethylene is processed below ca. 140°C, weak, unacceptable joints result. For this study a series of joints of decreasing strength were prepared by processing the polymer at temperatures below 140°C with the exception of the weakest joint which was prepared as described below. The scanning electron micrographs in Figure 1 show the cupric oxide surfaces after the branched polyethylene had been peeled away. Figures 1A through 1C represent the metal oxide surfaces for joints of increasing strength ranging from 1A termed a failure, to 1C called marginally strong. In particular, note the near disappearance of tooling grooves in Figure 1A compared to 1C. The weakest joint (Sample 1, Figure 1A) was prepared by hand buffing the copper oxide surface prior to forming the joint above 140°C. Buffing the surface not only redistributed the CuO particles so that the machined grooves were nearly filled, but also destroyed the typical needle-like appearance of the unbuffed surface (Figure 2A).[8] This perturbation of the surface in Sample 1 agglomerated the CuO fibers and produced a comparatively porous surface layer (Figure 2B). In contrast to this behavior, the surface of the strongest joint depicted in Figures 1C and 2A (Sample 3) clearly shows each lathe groove and only a slight massing of fibers. Sample 3 was prepared just below the proper processing temperature. The oxide layer in Sample 2, a joint of intermediate strength, was formed near 100°C. Apparently at this inadequate molding temperature application of pressure to the viscous polymer in order to fill the mold disturbs the Ebonol surface in a manner similar to the buffing operation (Figure 1B). The fourth sample was prepared according to the approved procedure.

The following experiments were performed on each sample: firstly, peel strength tests were made where the force required to strip a one-inch width of polyethylene from the oxide layer was measured; secondly, scanning electron micrographs were obtained for each surface after peeling; and thirdly, differential scanning calorimetry (DSC) was used to determine the

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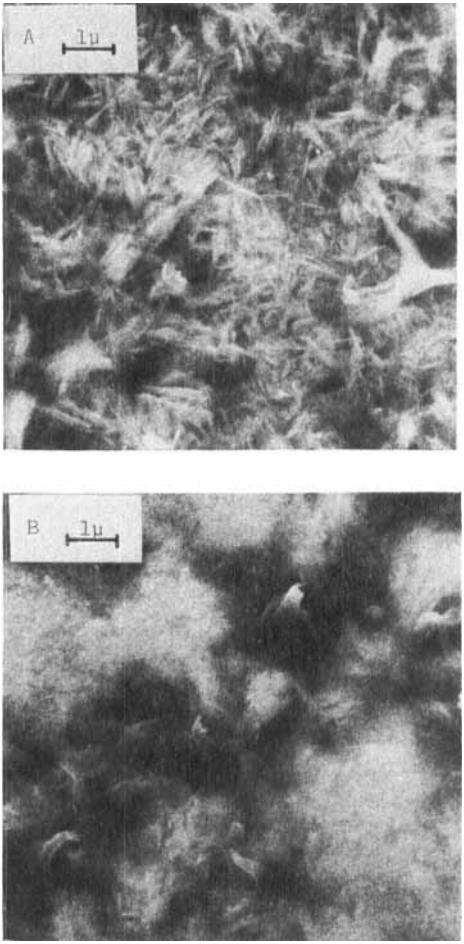

FIGURE 2 (A) Scanning electron micrograph of a copper oxide surface which would give a satisfactory joint under proper molding conditions. (B) A similar micrograph of the surface which has been buffed as in Sample 1, or Figure 1A.

amount of branched polyethylene adhering to the metal oxide surface after each peel test. The quantity of polymer, x, sticking to the CuO fibers was estimated from a comparison of the apparent heat of crystallization, $^*\Delta Q_c$, for the material scraped from approximately 0.60 in^2 of each copper block to the measured $^*\Delta Q_c^m$ of a known mass, M, of branched polyethylene (28μg); namely,

$$x = \frac{^*\Delta Q_c}{^*\Delta Q_c^m} M \qquad (1)$$

RESULTS AND DISCUSSION

The peel strengths range from 5 to 60 lbs. per inch of width for Samples 1 through 4. A comparison of the surface textures of the branched polyethylene which has been peeled away from the copper oxide is shown in Figure 3. The top micrograph (Figure 3A) is of Sample 1, which exhibited the poorest joint strength, 5 lbs. The middle picture (Figure 3B) is of Sample 2, which showed an improvement in peel strength, 13 to 21 lbs. The micrograph at the bottom of Figure 3 is of Sample 3, which was stronger than either of the preceding samples (26 to 40 lbs.). Sample 4, in which polyethylene necked and yielded with a test strength between 45 and 60 lbs., is not shown. In studying Figure 3, one observes an increasing surface roughness accompanies each improvement in joint strength.[9] Further, the almost featureless texture of the weakest joint (Figure 3A) results from essentially the same mode of failure as has occurred for the stronger joint (Figure 3C) except for the difference in the magnitude of the surface roughness. However, if the surface of Sample 1, the weak joint, is magnified 10 times more than the picture of the surface of Sample 3, the strongest joint, nearly identical surface textures, result for Samples 1 and 3, as illustrated in Figure 4. It should be noted that the peel strength of Sample 3 is almost an order of magnitude larger than Sample 1. Thus, in our experiments, the strength of a joint correlates with the polyethylene surface roughness.[8]

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In this study we shall consider two questions concerning this observation: (1) What is the relationship between the peel strength and surface roughness? and (2) What factors determine the degree of coarseness for the surface texture of polymer after peeling? It turns out that some mechanical aspects of the first question are very similar to those for the strength of rubber-reinforced solid plastics[10,11,12] in that the criterion of strength is not determined by the maximum stress that the polymer can withstand (yield stress), but by the amount of the total strain energy that is expended before the break,[13] including a large amount of work done during elongation (plastic deformation). Figure 5 shows our data on the force required to peel a one inch wide strip of polyethylene against the amount of polyethylene left on 1.00 in^2 of copper oxide surface after peeling. The peel strength is influenced by many factors. Besides the energy to break a unit area of the bonded surface, a strain energy in bending the strip of polymer as it is peeled must also be included. Since both processes involve large deformation, the rate dependence of the strength in the plastics is expected. For our study, only one kind of plastic is used; the rate of peeling and the geometry of the plastic strip being peeled are kept identical. The main variable in our case is, therefore, the energy required to create new surfaces. Consider a simplified peeling experiment as shown in Figure 6 in which a strip of plastic h inch wide

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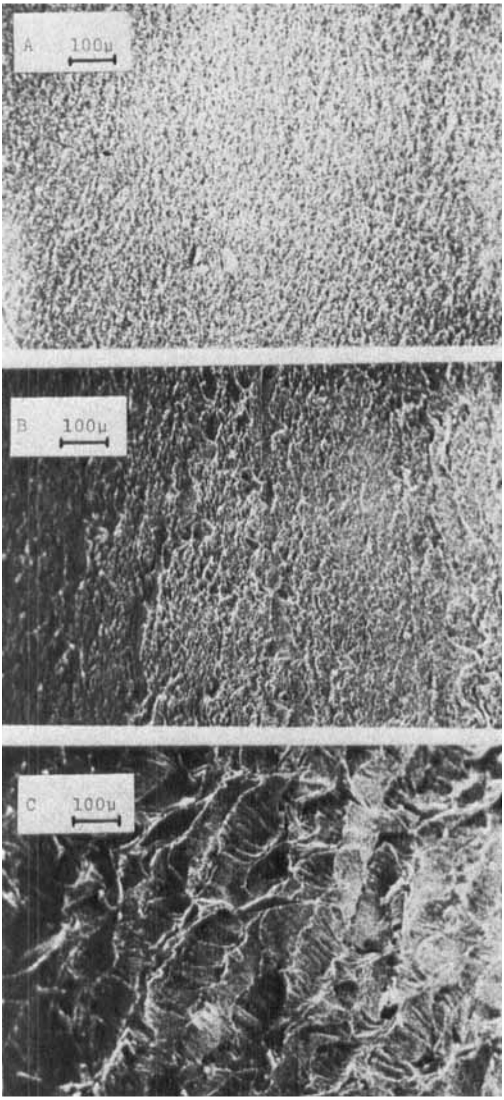

FIGURE 3 Electron scanning micrograph at same magnification of peeled polyethylene surfaces, (A) for Sample 1. (B) Sample 2, and (C) Sample 3. The progressively coarser surface texture corresponds to the increasing peel strength.

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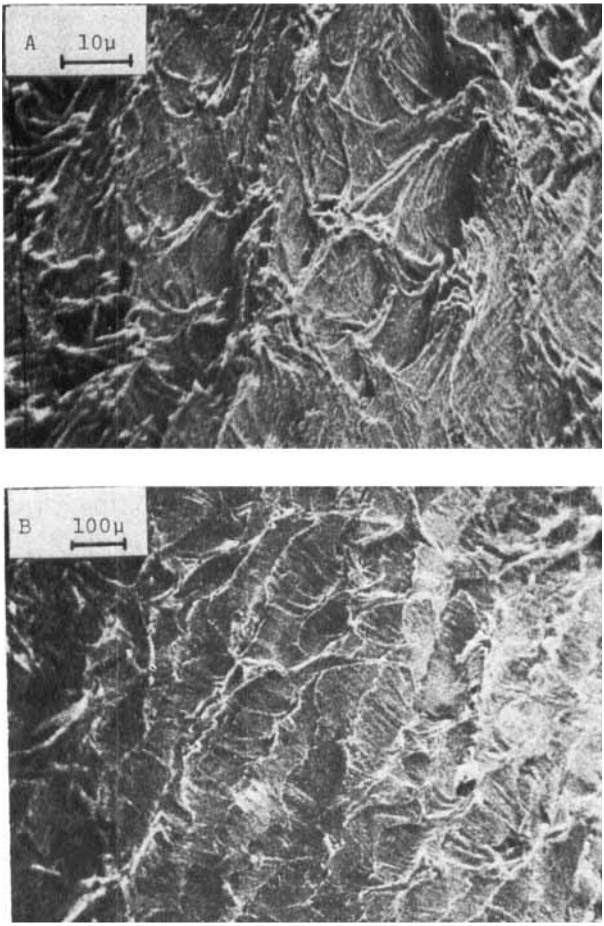

FIGURE 4 Electron scanning micrographs of two surfaces shown in Figure 3A and Figure 3C, now shown at different magnifications. The two surfaces now appear similar.

is peeled ΔC inches off the surface of the metal block and the force required during the peeling process is F pounds. The work, ΔW, done by the force during this process is

$$\Delta W = F\Delta C \text{ inch-pound} \tag{2}$$

where the effect of energy required for bending the strip is considered to be

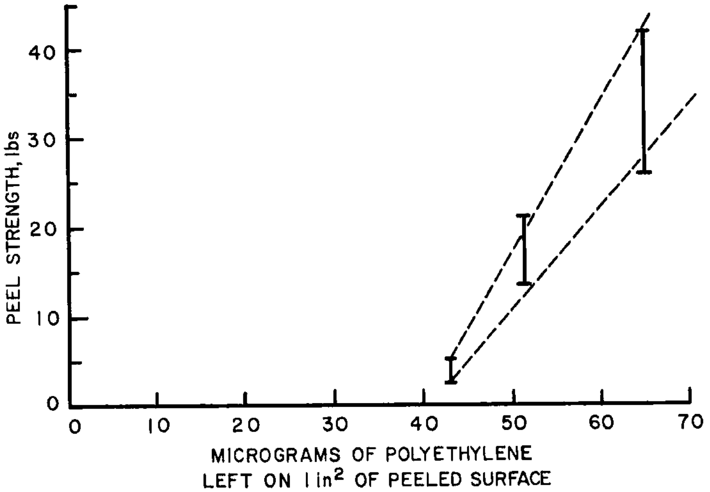

FIGURE 5 Peel strength vs. the amount of polyethylene left on one square inch of copper oxide surface.

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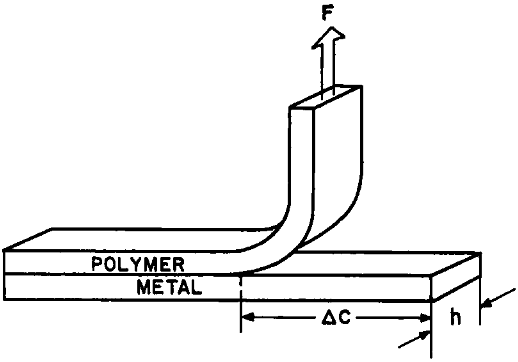

FIGURE 6 Schematic representation of the peel test.

much smaller than the energy for breaking the joint. It follows, then, that the energy, T, required to produce a unit area of the peeled surface, is

$$T = \frac{\Delta W}{h \Delta C} = \frac{F}{h} \text{ inch-pound/in}^2 \tag{3}$$

Thus, the ordinate of the graph in Figure 5 is numerically identical to the energy required to produce a square inch of peeled surface, since in this case h = one inch. The energy required to create such a surface often exceeds, by some orders of magnitude, the theoretical maximum limit of creating a smooth new surface by considering the breaking of molecular or chemical bonds.[14] For the case of propagation of a crack in a polymeric solid, this large excess energy required to create the failure was shown by Berry[14] to arise from the strain energy dissipated in the bulk during plastic deformation, particularly in the vicinity of the tip of the crack. We have also shown[12] that the rubber reinforcement for the matrix of solid polymer is accomplished through a greater plastic deformation of the solid component in the vicinity of the maximum stress concentration near the interface with the rubber particles. In either case, the mechanical advantage is accomplished in a system which requires a greater dissipation of mechanical energy prior to the actual parting of the two surfaces. When this viewpoint of the "strength" of polymer solids is taken into consideration, the results, shown in Figure 5, seem completely reasonable, i.e. when a greater amount of polyethylene is involved in plastic deformation, a proportionately greater amount of energy is required. Thus we have the answer to the first of the two questions, i.e. the reason for the correlation between the peel strength and the texture of the peeled polyethylene surface.

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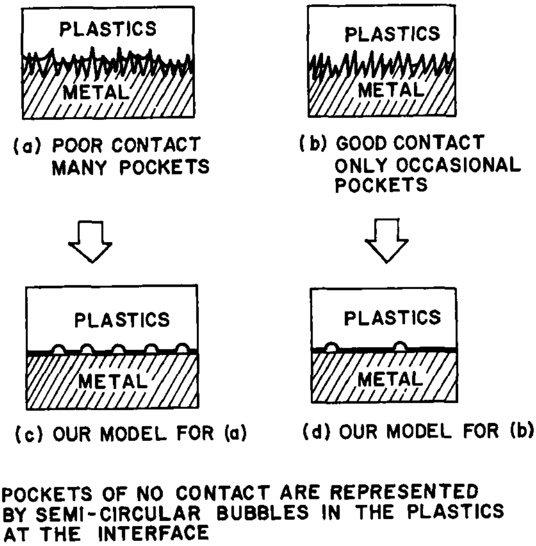

FIGURE 7 Model describing two cases of interfacial contact.

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We now wish to consider the second question, which was what relationship exists between the quantity of deformed polymer and the degree of oxide–polymer contact? In our study this quantity of drawn polymer was found to increase as the amount of "contact" was improved. We shall attempt to explain this observation by proposing a mechanical model to represent stress distribution near the interface. The concept of a good or a poor contact from the adhesion standpoint by Schonhorn[1] is schematically shown in Figure 7, A and B, respectively. Figures 7C and D are our models for such joints, in which the absence of the contact is represented by the population of the semi-circular voids per unit dimension of the interface. (For simplicity, we consider the 2-dimensional case. The 3-dimensional model would be only numerically different in certain quantities involving the stress concentration, but the basic nature of the problem remains unaltered). The distribution of stress for such a model, shown in Figure 8B, can be considered similar to that around a hole in a plate of plastics, shown in Figure 8A, except for the dilational stress arising from the rigidly fixed interface in the former.

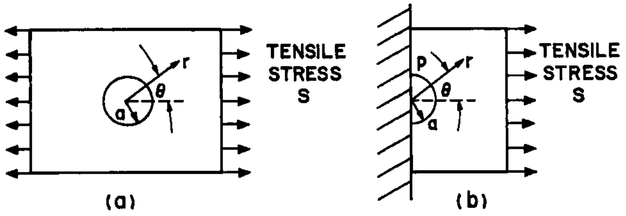

FIGURE 8 Stress distribution around a void in the solid body (A) is shown to be similar to the distribution at the adhesive interface with an imperfect contact such as described in Figure 7.

This dilatational effect will no doubt affect the viscoelastic properties of the polymer. However, in this study the effect of stress distribution from the geometry around a hole or holes is probably the predominant factor. The stresses, if we ignore this additional dilatational stress, is given by[15]

$$\sigma_r = \frac{S}{2}\left(1 - \frac{a^2}{r^2}\right) + \frac{S}{2}\left(1 + \frac{3a^4}{r_4} - \frac{4a^2}{r_2}\right)\cos 2\theta \tag{4a}$$

$$\sigma_\theta = \frac{S}{2}\left(1 + \frac{a^2}{r^2}\right) - \frac{S}{2}\left(1 + \frac{3a^4}{r^4}\right)\cos 2\theta \tag{4b}$$

$$\tau_{r\theta} = -\frac{S}{2}\left(1 - \frac{3a^4}{r^4} + \frac{2a^2}{r^2}\right)\sin 2\theta \tag{4c}$$

where σ_r and σ_θ are the normal stresses in r and θ directions, and $\tau_{r\theta}$ is the shear stress in the θ direction on the plane normal to the r direction. The maximum stress occurs at the edge of the circular hole, designated by p in Figure 8B, for which $r = a$, $\theta = \dfrac{\pi}{2}$, $\sigma_r = 0$, so that

$$\sigma \text{ at } p = \sigma_\theta\left(\theta = \frac{\pi}{2}, r = a\right) = 3S \tag{5}$$

i.e., the stress at $(a, \pi/2)$ is three times that of the overall tensile stress. The concentration of stress near this point of maximum stress is a rapidly decaying function of r, so that at $r = 2a$, the stress is only 20% greater than the overall tensile stress S, as illustrated in the upper part of Figure 9.

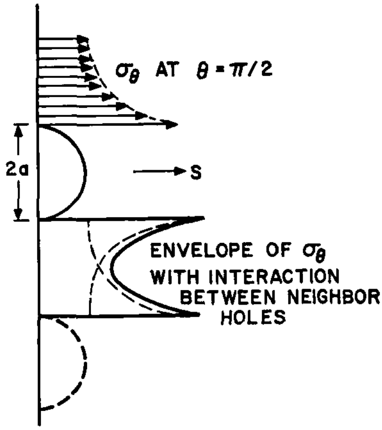

FIGURE 9 Stress concentration around a hole (the region of no contact) as influenced by an adjacent hole.

The sharply decreasing stress concentration means that the maximum stress is not substantially increased by the presence of a neighboring hole unless it is situated extremely close. To illustrate this point the above statement can be reworded as follows: Even when the holes are separated by their own diameter, the maximum stress will rise only by $0.2S$ or less than 7% of the total magnitude. Thus, from this model we draw the conclusion that the proximity among the holes, representing the poor contact between plastics and the metal, contributes little to raising the maximum stress concentration. Thus, we must look for some other features of stress distribution that are altered by changing the distance between the holes. Let us consider the stress component in the direction of the tensile stress, S, along the line $r = r$, $\theta = \pi/4$, as shown in Figure 10. Such a stress, defined as σ_x here, can be derived from Eqs. 4a, b, and c by using the relationship

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$$\sigma_x = \sigma_r \cos^2\theta + \sigma_\theta \sin^2\theta - 2\tau_{r\theta}\sin\theta\cos\theta \qquad (6)$$

so that

$$\sigma_x\left(r = r, \theta = \frac{\pi}{4}\right) = \frac{S}{2} + \frac{S}{2}\left(1 - \frac{3a^4}{r^4} + \frac{2a^2}{r^2}\right) \qquad (7)$$

σ_x, thus, is a monotonically increasing function of r, for all $r > a$, approaching the nominal tensile stress, S, as r is increased, *only if one hole is considered.* If a neighboring hole exists, the situation becomes quite different, as shown in Figure 10. Where the two lines $\left(r = r,\ \theta = \frac{\pi}{4}\right)$ and $\left(r = r,\ \theta = -\frac{\pi}{4}\right)$ for the adjacent holes meet, designated by point q, the stress now exceeds that of the overall stress. This means that the failure could start at q. (We have considered only interactions between nearest neighbors, others are expected to be less important in practice.) *This means that the polymer between this point q and the interface can be involved in plastic deformation,* and it is obvious from Figure 10 that this distance t is related to the distance between the holes, δ, by $t = 0.5\delta$. Alternately, the failure pattern could be

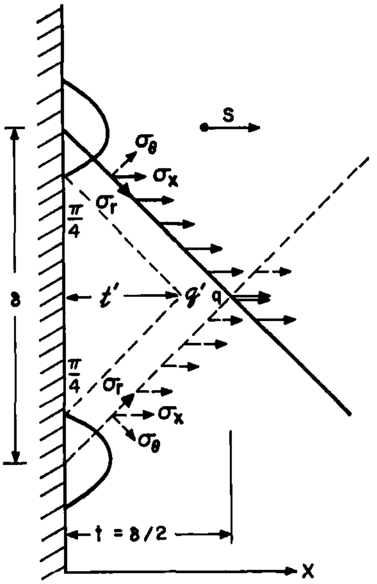

FIGURE 10 Schematic diagram describing the mechanical model for the thickness of the weak boundary layer near the interface.

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explained with plasticity theory.[16] We note in Equation 4 that the maximum principal shear, which is one half of the difference between the two principal stresses, is maximum at the edges $\left(\theta = \pm\frac{\pi}{2}\right)$ of the circular hole. Furthermore, the planes on which the maximum principal shear acts are inclined at ±45° from the interface. Thus, according to Tresca's yield criterion,[15] the polymer will fail by developing slip planes from the edges of the circular holes at the angles of ±45° (Figure 10). The distance t between the interface and the first intersection, q, of any two slip planes is $t = (\delta - 2a)/2$, which is approximately equal to $t = \delta/2$ if $2a \ll \delta$.

We have now shown, at least within the realm of consistency for this model, that the nature of contact between the polymer and the oxide surface determines the amount of polymer to be involved in plastic deformation before the joint is completely separated, and the strength of the joint is determined by the total energy involved in so deforming the polymer. To examine the plausibility of this model, let us now consider what sort of dimensions are being considered here. Let us look at the picture of the oxide surface shown in Figure 2A. The typical dimension between the needles seems to be in the order of 0.1 microns. Thus, for marginally strong joints such as the Samples 2 and 3, δ may very well be taken to be also 0.1 microns. This will give the thickness of the layer that we considered in Figure 9, to be $t \approx 0.05$ microns, or 500 Å. The amount of polyethylene left on the surface for Samples 2 and 3 was estimated from differential scanning calorimetry to be between 50 and 65 micrograms per square inch of surface. By assuming the density of the plastics to be about 1 gm/cm^3, we obtain the initial thickness of polyethylene which went through the plastic deformation (drawing) ranges from 800 to 1100 Å. The general agreement seems to indicate that our proposed model may be a reasonable one.

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CONCLUSIONS

For some adhesive joints where the main difference is the degree of contact at the interface, failure occurs not at the interface, but some distance away in the polymer itself, involving most of the material in this layer in local plastic deformation. The strength criterion for such a joint is the energy of deformation and not the maximum tensile stress in the material. In addition, even in the weakest joint prepared for this study the polyethylene failed cohesively.

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