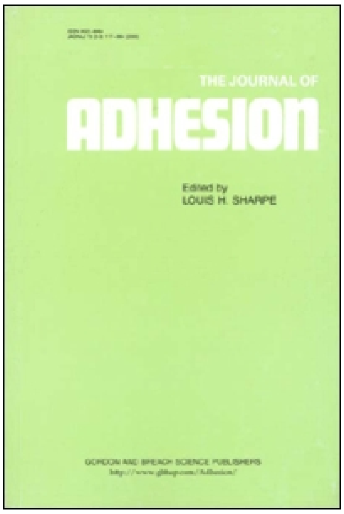

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Quasi-Static Adhesive Fracture

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Quasi-Static Adhesive Fracture

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The concept of quasi-static crack propagation is used in the present paper to study quantitatively the effects of environmental fluids on fracture in adhesive joints. The mechanisms and mechanics of environmental adhesive fracture under rising loads are discussed. Two types of cracking behaviour were observed. (1) When the dissolution or the "surface energy reduction" mechanism prevailed, the fracture toughness of the adhesive joint in the environment was reduced. (2) However, when environment-enhanced crazes were formed in the adherend at the crack tip region, the local fracture toughness of the adhesive joint would be increased. But cracking was usually unstable so that crack velocities were not readily measurable.

Except in so far as the adhesive surfaces may have considerable effects, the fracture toughness of an adhesive joint is independent of the specimen geometries used in the present work. Also, the variation of fracture toughness with crack velocity for an Aluminum/Araldite joint in a carbon tetrachloride solution is reported.

1. INTRODUCTION

For any structural adhesive joint it is important and necessary to know how the fracture toughness† (R), defined as the work required to create unit area of crack surface, varies with temperature (T), glue thickness (t), crack front velocity ($\dot{L}$) and absorption of appropriate hostile environments. Relations between R and these variables may be established independently from suitable experiments using the quasi-static crack propagation method of Gurney and Hunt.[1]

Some important experimental work has been done by Ripling *et al.*[2, 3] and Mostovoy and Ripling[4, 5] on the fracture behaviour of aluminum–epoxy–aluminum adhesive joints. They[2] have found that over a wide range

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† R is a symbol used in Gurney and Hunt and is essentially equal to G_c, the critical strain energy release rate. For quasi-static cracking to occur, $ER = bK^2$, where E is Young's modulus, K, stress intensity factor, and $b = 1$ for plane stress and $1-\gamma^2$ for plane strain. γ is Poisson's ratio.

of bond thickness (0.002 to 0.4 inch) and width (0.25–1.0 inch) dimensions, the fracture toughness values of these adhesive joints are reasonably consistent. In contrast, the roles played by postcure temperature and absorption of moisture contents in affecting the fracture toughness of the epoxy adhesive joints are more significant. Mostovoy and Ripling[4] showed that R of the joint could be increased by as much as 4 times when the postcure temperature was increased from 180 to 350°F. They have also demonstrated two significant effects caused by the absorption of moisture at the crack tip. First, in a rising load experiment, the moisture can considerably increase the fracture toughness of the adhesive joint; and second, in constant G (equivalently R) experiments, with G less than G_{1c} for unstable crack extension, the crack grows slowly with a velocity dependent on the applied G-level.

Despite these definitive contributions[2-5] to the understanding of fracture mechanisms and mechanics in aluminum/epoxy adhesive joints, still much more work is needed for other adhesive/environment systems. For example, the mechanisms of fracture and the variation of fracture toughness with absorption of appropriate environmental fluids at the crack tips would vary with the kind of adhesive joints used. It is therefore the purpose of the present work to evaluate these effects and to study the possible mechanisms controlling fracture in some typical adhesive joint-environment systems. Some experimental results showing the $R(\dot{L})$ relationship for an aluminum/Araldite adhesive joint in carbon tetrachloride are also presented.

In the present investigation, adhesive joints were made using Perspex, aluminum and polycarbonate (Makrolon) as adherends and Araldite (Ciba) and Tensol Cement (I.C.I.) as adhesives.

2. THEORY OF QUASI-STATIC CRACK PROPAGATION

Consider for simplicity two halves of adherends of rectangular beam-like geometry stuck together with an adhesive except for a short central crack having a nominal surface area of one side (A), under the action of two equal and opposite self equilibrating sets of forces (X), with relative normal displacement (u) (see inset of Figure 3) the work equation of mechanics† gives

$$X\,du - d\Lambda = R\,dA + dK \tag{1}$$

Λ is the strain energy function, K the kinetic energy function, and $A = tL$, where t is the width of the joint and L the crack length.

For quasi-static cracking processes, dK is small compared with other quantities and when residual stresses are absent, it may be obtained that Λ is only a function of (X, A) or (u, A). Thus, from Eq. (1) the following relations

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† In the expression (1) given, time independence of material properties is assumed.

are derived:

$$X = \left(\frac{\partial \Lambda}{\partial u}\right)_A ; \; R = -\left(\frac{\partial \Lambda}{\partial A}\right)_u . \tag{2}$$

The fracture toughness (R) defined in the second of Eq. (2) is a material property of the adhesive joint being independent of the adherends except in so far as the adherend surface may affect it.

A concept of crack spreading in the quasi-static regime can be found in the work of Gurney and co-authors[1, 6-8] in which a very simple irreversible work area method of determining fracture toughness of materials in elastic structures is discussed. In general, the Gurney-method for fracture analysis is useful as long as the cracking experiments are continuous and stable so that simultaneous measurements of R and corresponding crack speeds are easily obtained. It should be noted that in conventional toughness testing where unstable specimen geometries are employed, cracking once started goes unstably with a "bang". These situations are schematically shown in Figure 1. The limiting speed of the quasi-static concept has been estimated in Gurney and Ngan[7] and found to correspond to one thousandth of the longitudinal wave velocity of the material.

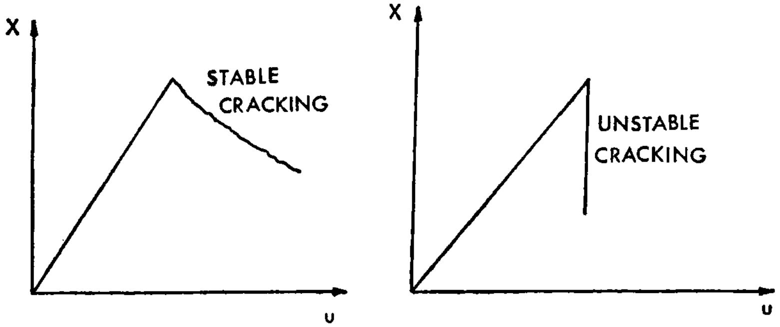

FIGURE 1 Schematic load-deflection (X–u) diagrams for stable and unstable cracking experiments.

3. EXPERIMENTAL

Fracture experiments on adhesive joints were performed using three different testpiece geometries commonly employed in Fracture Mechanics studies. These specimens with labelled dimensions are shown in Figure 2a–2c and are known as the edge-crack-line loaded (ECLL) specimen, the double-ended cantilever beam (DCB) testpiece and the Outwater double torsion (D.T.) specimen. The idea of using various test geometries is to see if the toughness of a given adhesive joint is truly geometry independent. In the present work, crack spreading in the following adhesive joints is discussed;

a) Aluminum/Araldite
b) Polycarbonate (PC)/Araldite
c) Polycarbonate/Tensole Cement
d) Perspex/Tensole Cement
e) Perspex/Araldite

To prepare a proper adhesive joint for the experiment, the surfaces of the two halves of adherends to be bonded together were roughened with 400 A emery cloth, cleaned and then suitably annealed so that any residual stresses could be removed. The adherends were then stuck together with the adhesive except for a short length which constituted a "natural" crack. Curing of the adhesive was allowed for at least 24 hours.

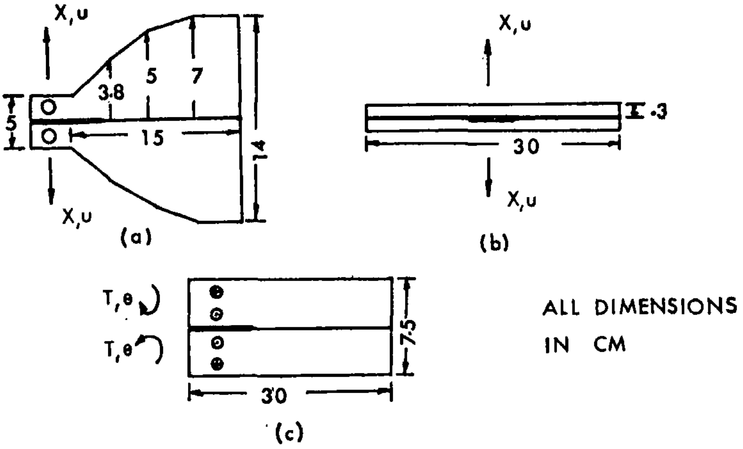

FIGURE 2 a) Edge crack-line loaded specimen
b) Double ended cantilever beam testpiece
c) Outwater double torsion specimen

The well-prepared specimens were tested in an Instron testing machine under pre-determined conditions. In the environmental fracture tests, little reservoirs were constructed on the specimens so that the crack tips were continuously in contact with the liquid. The fracture toughness values of the adhesive joints were calculated using the Gurney-method and the corresponding crack speeds determined with the aid of a travelling microscope.

4. RESULTS AND DISCUSSION

4.1. Effect of specimen geometry on toughness measurement

The experimental results on the various adhesive joints were tabulated in Table I. The testing conditions were 48% relative humidity, 22–23 degrees

Celsius and an air environment. The crack speed was roughly 10 cm/min. In general, R is truly a parameter of the adhesive joints and is independent of specimen geometry. Perhaps, the only exception is the PC/Araldite adhesive joint which shows considerable scatter in R between the ECLL and the DCB specimens. A possible reason for this difference is obvious because polycarbonate and Araldite do not form good adhesive joints. Reproducibility is therefore difficult to achieve.

TABLE I

Variation of toughness of adhesive joints with specimen geometry (Crack speed $\approx$ 10 cm/min)

Adhesive joint	Specimen geometry	R (kg/cm)
Perspex/Araldite	ECLL	0.14–0.16
	DCB	0.13–0.15
	DT	0.15–0.18
Perspex/Tensol Cement 6	ECLL	0.30–0.31
	DCB	0.29–0.30
	DT	0.30–0.31
Perspex/Tensol Cement 7	ECLL	0.57
	DCB	0.55–0.56
	DT	0.59
PC/Tensol Cement 6	ECLL	0.30–0.32
	DCB	0.32
PC/Araldite	ECLL	0.05–0.08
	DCB	0.07–0.12
Aluminum/Araldite	ECLL	0.72
	DCB	0.70–0.75

4.2. Mechanisms and mechanics of crack extension in hostile liquid environments

When environments are absorbed at the crack tip region, the adhesive (as well as the adherend) may be dissolved, swollen or unchanged, all resulting in considerable changes in the crack tip stress–strain fields and effective fracture toughness of the joint. An analysis on the variation of the local stress intensity factor (K) and R with absorption with environmental fluid at the crack tip region has been attempted in Mai.[9] However, the individual effect of environment absorption on K or R cannot be completely assessed.

In general, under rising load experiments, two types of cracking behaviour in the adhesive joints used in the present work were observed. In the type I behaviour, the fracture load was reduced and the effective fracture toughness of the adhesive joint decreased. Cracking was stable and continuous. As for the type II behaviour, the environment would cause both the fracture load and the toughness to increase significantly. Cracking, however, was usually unstable. Specific examples are given below.

Type I cracking behaviour This cracking behaviour under rising load is best exemplified by a Perspex/Araldite joint in the presence of a carbon tetrachloride solution. Figure 3 shows a load-displacement ($X-u$) diagram of a peeling experiment performed on the Araldite joint using a DCB specimen with bond width of 26 mm. The adhesion toughness of the joint was reduced to 0.063 kg/cm when compared with that in a reference environment (air) at room conditions (0.147 kg/cm). Since carbon tetrachloride does not usually cause dissolution or appreciable swelling in epoxy within the time scale of our experiments, and since examination of the Perspex adherend surfaces shows no traces of crazing,† the reduction in toughness of the joint may be reasonably attributed to a possible "surface energy reduction" mechanism due to

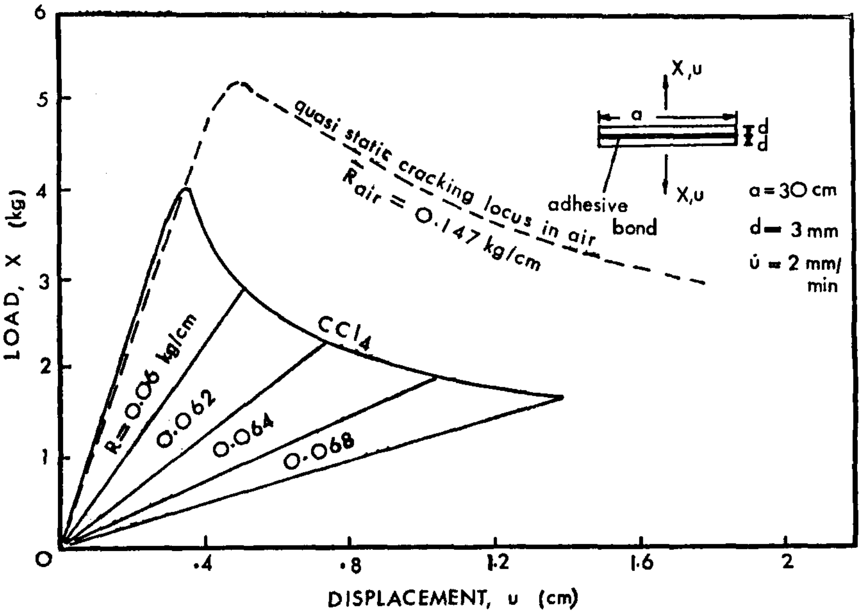

FIGURE 3 Splitting of a Perspex/Araldite joint in a carbon tetrachloride solution.

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† The absence of crazes is not uncommon if the preferential reduction of the adhesive bond strength is greater than the crazing stress necessary for Perspex. Cracking along the line of adhesion would therefore occur before crazes could be developed in the adherend surfaces.

adsorption of the environments. This is analogous to the mechanisms controlling liquid-metal embrittlement.[10, 11]

Another example was the case of cracking a Perspex/Araldite adhesive joint in tap water. This further supports the idea of a surface energy reduction mechanism because water does not swell or dissolve either the adherend or the adhesive. The reduction in fracture toughness of the joint must therefore be a direct consequence of water in lowering the interfacial bond strength by adsorption.

Note that type I cracking behaviour also exists in cases where dissolution of the adhesive occurs.

Type II cracking behaviour Figure 4 records the experimental results on a Perspex/Tensol Cement 6 joint using a DT specimen of 6 mm bond thickness. The environment used was also carbon tetrachloride. Obviously, the cracking load was increased when CCl_4 was added to the crack tips. This can be

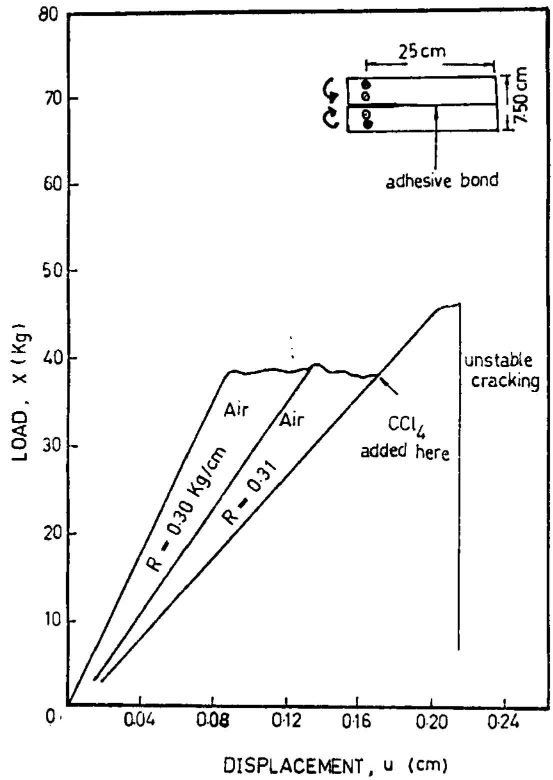

FIGURE 4 Cracking of a Perspex/Tensol Cement 6 joint in air and in carbon tetrachloride.

compared with the lower fracture load required for quasi-static cracking in air. In the DT specimen, a higher cracking load would imply a higher fracture toughness value. This has been proved in Kies and Clark[12] and in Mai and Gurney.[13] It is not entirely clear why the toughness is substantially increased in this example. However, a plausible explanation is given here. Although CCl_4 does not swell or dissolve the adhesive (i.e. Tensol cement) its hostile action on the Perspex adherends is definite. Crazes in Perspex were known to form under the combined action of applied stress and environment in numerous published work.[14-19] When the crazes were nucleated at the crack tip, the net effect was to relieve the high stresses that would otherwise have been set up. The crack tip was therefore apparently blunted (in fact, this was multiple branching in a small volume of the crack tip material)—this would cause a considerable drop of the local stress intensity factor and would facilitate the accumulation of strain energy in the crack tip material.

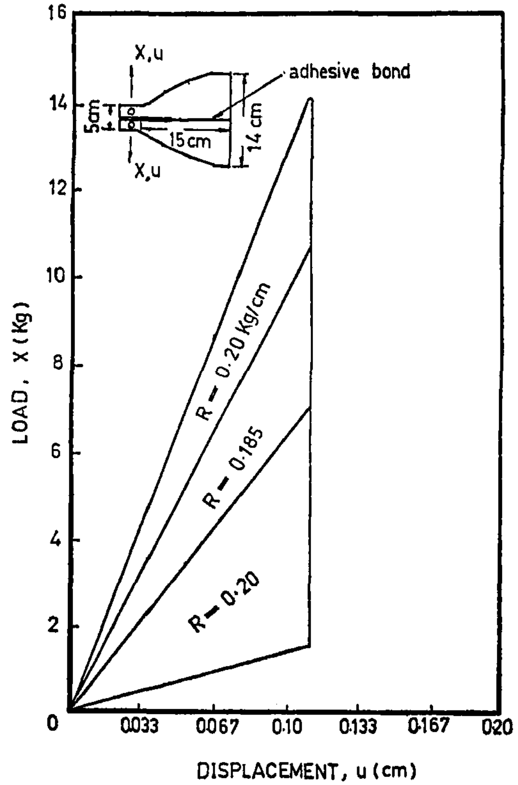

FIGURE 5 Splitting of a Perspex/Tensol Cement 7 joint at constant displacement in carbon tetrachloride.

In the present adhesive joint/environment system, crazes indeed were developed in the Perspex adherends† at the crack tip region. Because of the stress-relief effect of these environment induced crazes, a higher than usual (i.e. in air) fracture load was required for cracking and a higher fracture toughness was apparently obtained. However, cracking was usually unstable so that the crack velocity was not measurable or cracking was so catastrophic that a considerable amount of kinetic energy was lost to the surroundings.

However, stable cracking in the Perspex/Tensol Cement joint is not entirely impossible. Figure 5 records such a stable cracking experiment in carbon tetrachloride at fixed grip condition (i.e. sustained deflection). The bond thickness was 6 mm. Note that if splitting is caused by a monotonically increasing load, the cracking is unstable. By allowing time for the liquid to penetrate to the crack tip region, stable splitting of the joint was found. The fracture toughness was reduced (0.20 kg/cm), corresponding to a low value of the crack front velocity. It was also observed that as the main crack front propagated to create new surfaces, advanced nucleations of minute cracks were generated ahead of the crack tip region.

When the aluminum/Araldite joint was cracked in water, this also displayed the type II behaviour. The toughness of the joint could be increased as much as 100% when the crack tips were moistened with water. However, because water certainly could not cause crazing in either the aluminum or the adhesive, it was not clear why and what would attribute to this fracture toughness increase phenomenon.

4.3. Crack velocity—fracture toughness relationship

For a given adhesive joint in a given environment and stress situation, it is useful to obtain some information showing the variation of fracture toughness with crack speed. As an example, consider Figure 6 which shows the R–$\dot{L}$ results for an aluminum/Araldite joint in carbon tetrachloride. It is obvious that the fracture toughness increases with crack velocity. It also appears that for crack velocities below 10^{-1} mm/s, a threshold fracture toughness of about 0.06 kg/cm exists.

Because CCl_4 does not induce either swelling or dissolution of the metal adherend or the Araldite, the reduction in fracture toughness must therefore be a direct consequence of "surface energy reduction" by the environment. This argument is further supported by the fact that the adhesive joints usually exhibit interfacial fracture.

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† Cracking along the adhesion does not occur via the "surface energy reduction" postulate probably because the adverse effect of CCl_4 on Perspex is more severe than on the Tensol Cement bond.

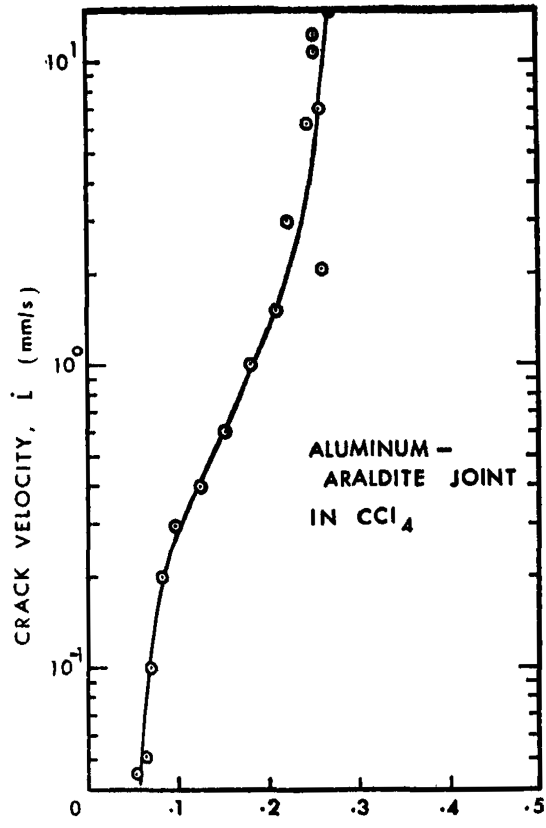

FIGURE 6 R-$\dot{L}$ relation for an Aluminum/Araldite joint in carbon tetrachloride.

4.4. Summary of results

Table II summarizes the variation of fracture toughness with various liquid environments for five adhesive joints studied in the present work. The toughness values shown correspond to crack speeds of about 10 cm/min.

It should be noted that in the PC/Tensol Cement joint, all cracking experiments in ethanol and carbon tetrachloride were not successful. These organic solvents were exceedingly detrimental to polycarbonate (e.g. the fracture toughness of polycarbonate in $CCl_4 \approx 0.1 \sim 0.2$ kg/cm) and resulted in failure of the adherends but not the bond.

TABLE II

Fracture toughness of adhesive joints in hostile environments (crack speed ≈ 10 cm/min)

Adhesive joints	R^* (kg/cm)	R_{air}/R^*	Environment
Perspex/Araldite	0.147		air
	0.063	9.0	water
	0.062	9.0	CCl_4
	0.20–0.30 (unstable)	0.5–0.74	ethanol
Perspex/Tensol Cement	0.570		air
	0.2–0.4	1.4–2.8	CCl_4
	0.70–0.80 (unstable)	0.7–0.82	ethanol
PC/Tensol Cement	0.30		air
	failure of adherend	—	CCl_4, ethanol
	unstable	—	NaOH
PC/Araldite	0.05		air
	0.012	4.5	CCl_4
	0.02	2.5	water
Aluminum/Araldite	0.72		air
	0.26	3.50	CCl_4
	0.85	0.82	water

4.5. Fracture morphology

Figure 7 shows in detail the mechanism of crack growth of an adhesive joint in environmental fluids. Interfacial cracks may be generated at the lower and upper interfaces simultaneously and then continue to extend along the entire specimen length. The adhesive bond may be sheared off. However, experiments also showed that under room conditions in air, the crack spread along one interface only. There are reasons controlling these modes of fracture.

If we assume R_x, R_{xy} and R_y as the fracture toughness of the adherend, the interface and the adhesive respectively; then for cracking along one entire interface, we must have the conditions

$$R_{xy} < R_y \quad \text{and} \quad R_{xy} < R_x. \tag{3}$$

For fracture of the adhesive joint in a hostile environment, all magnitudes of these three quantities are effectively changed. If the condition

$$R_x < R_{xy} < R_y \tag{4}$$

prevails, we expect fracture to occur across the adherend. The best example seen from experiments reported here is the PC/Tensol Cement adhesive joint when fractured in CCl_4. On the other hand, if we have

$$R_y < R_{xy} < R_x \quad \text{or} \quad R_{xy} < R_y < R_x \tag{5}$$

at any instant during crack spreading we will have intermittent fracture along the interfaces and shear fracture across the adhesive. This is the common way in which most structural adhesive joints fail in a hostile environmental fluid.

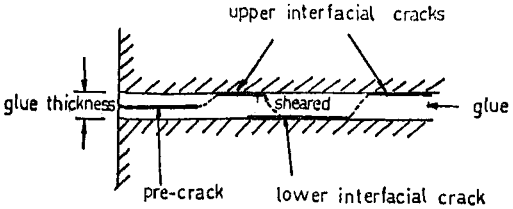

FIGURE 7 Cracking mechanism of an adhesive bond in the presence of environmental fluids.

5. CONCLUSIONS

When characterizing the resistance to cracking of an adhesive joint it is important and possible to determine the effects of absorption of environmental fluid at the crack-tip region on the fracture toughness of the joint.

It has been shown that in general two types of cracking behaviour would be obtained. One type results in the lowering of fracture toughness of the adhesive joint by the environment through a dissolution or surface energy reduction mechanism; the other causes the fracture toughness to increase because the crack tip is apparently blunted by crazes formed in the adherend.

While the fracture toughness of an adhesive joint does not depend on the test configuration, its dependence on crack velocity is significant. Continuing experiments are being performed in order to obtain the R–$\dot{L}$ relationship for some other common adhesive joint/environment systems. It is expected that these results will be reported soon.

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