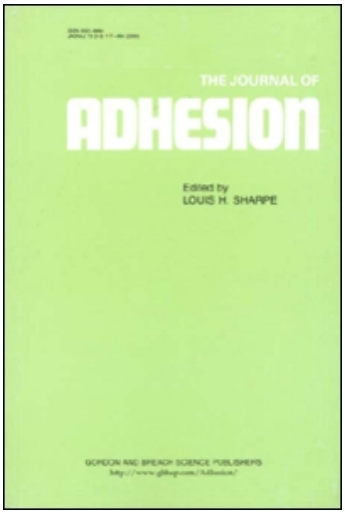

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The Fatigue and Durability Behaviour of Automotive Adhesives. Part I: Fracture Mechanics Tests

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The Fatigue and Durability Behaviour of Automotive Adhesives. Part I: Fracture Mechanics Tests

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A fracture mechanics approach has been successfully used to examine the cyclic fatigue behaviour of adhesively-bonded joints, which consisted of aluminium-alloy or electro-galvanised (EG) steel substrates bonded using toughened-epoxy structural paste-adhesives. The adhesive systems are typical of those being considered for use, or in use, for bonding load-bearing components in the automobile industry. The results were plotted in the form of the rate of crack growth per cycle, da/dN, *versus* the maximum strain-energy release rate, G_{max}, applied in the fatigue cycle, using logarithmic axes. Of particular interest was the presence of a threshold value of the strain-energy release rate, G_{th}, applied in the fatigue cycle, below which fatigue crack growth was not observed to occur. The cyclic fatigue tests conducted in a relatively dry environment of 23°C, and 55% ; RH were shown to cause crack propagation at far lower values of G_{max} compared with the value of the adhesive fracture energies, G_c, which were determined from monotonically-loaded fracture tests. Cyclic fatigue tests were also conducted in a "wet" environment, namely immersion in distilled water at 28°C. The "wet" fatigue tests clearly revealed the further significant effect an aggressive, hostile environment may have upon the mechanical performance of adhesive joints, and highlighted the important influence that the surface pretreatment, used for the substrates prior to bonding, has upon joint durability. The development and standardisation of "wet" fatigue tests may provide the basis for a very effective accelerated-ageing test.

Keywords: Aluminium alloy; automotive applications; durability; electro-galvanised steel; fatigue; fracture mechanics; structural adhesives; surface pretreatments

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1. INTRODUCTION

The present research is particularly directed towards adhesives for automotive applications. Adhesives are currently used in many areas in the manufacture of automobiles, but almost always either as basically sealant materials or in noncritical secondary structures. So far the use of adhesives in truly structural applications has been very limited. A major reason for this has been a concern about the fatigue and durability behaviour of bonded, structural components over the expected lifetime of the vehicle. Since the adhesive joints must perform satisfactorily under service conditions, which include dynamically applied loads and exposure to hostile environments such as water, petrol, other organic solvents, etc. and, in many instances, combinations of these conditions, may be experienced.

It is, therefore, of prime importance for the adhesives technologist to be able to develop and recommend "adhesive systems" (*i.e.* the substrate/surface pretreatment/adhesive) which will possess an adequate service life under the operating conditions which are to be experienced by the bonded structure. This, in turn, leads to the need to understand the mechanisms of failure and to develop test methods (i) for developing and selecting adhesive systems, (ii) for quality assurance, and (iii) for predicting, quantitatively, the expected service life.

Dynamic fatigue is the phenomenon of failure or fracture of a material, joint or structure under repeated or oscillatory loading [1]. The importance of dynamic fatigue is that under fluctuating loads joints will fail at stress levels much lower than they can withstand under monotonic loading or under static (*i.e.* creep) loading. Further, it is well established that the mechanical performance of adhesive joints may be adversely affected when exposed to aqueous environments, especially at an elevated temperature [1]. Therefore, subjecting adhesive joints to dynamic fatigue loads, whilst being immersed in water, is potentially a very demanding test environment, but one which is frequently encountered in a bonded structure.

However, apart from representing a real-life environment, it is also possible that the use of dynamic loading will accelerate the kinetics of the attack upon the joint by the ingressing water. Such an effect would be of some importance, since other methods of accelerating the attack by water upon joints often involve greatly increasing the temperature, or applying unrealistically high static loads.

These methods may drastically change the basic mechanism of joint failure, rather than merely accelerate the mechanism which is observed to occur during the service-life of the bonded structure [1].

As with other materials, the fatigue behaviour of adhesives and adhesive joints has been successfully studied employing a continuum fracture mechanics approach [1–9]. The early work by Mostovoy and Ripling [2] clearly established the validity of using a linear-elastic fracture-mechanics (LEFM) approach for describing the fatigue crack growth behaviour when bonding aluminium-alloy substrates using a range of epoxy-based adhesives. They employed a tapered double cantilever-beam (TDCB) joint specimen, see Figure 1, and conducted the tests under nominally mode I (tensile-opening) cyclic loading and measured the rate of crack growth per cycle, da/dN, as a function of the applied range of strain-energy release-rate, ΔG, that was imposed, where:

$$\Delta \mathbf{G} = \mathbf{G}_{\max} - \mathbf{G}_{\max} \tag{1}$$

and G_{max} is the maximum and G_{min} is the minimum value of the strain-energy release-rate applied per cycle. Firstly, they observed that, as for many other materials, over much of the range of experimental data the crack growth rate may be expressed by:

$$\frac{\mathbf{da}}{\mathbf{dN}} = \mathbf{A}_f \Delta \mathbf{G}^q \tag{2}$$

where A_f and q are constants. Secondly, their studies revealed that the relationship between da/dN and ΔG was actually sigmoidal in shape. Crack growth rates were found to decrease to very low values as ΔG approached some limiting threshold value, G_{th}, and to increase to very high values as ΔG approached the typical value of the adhesive fracture energy, G_c, for crack growth under short-term monotonic loading conditions.

The aim of the present work was to study the dynamic fatigue behaviour of joints which consisted of aluminium-alloy or electro-galvanised (EG) steel substrates bonded using epoxy-based structural paste-adhesives. The adhesives chosen were typical of those used in the automotive industry. One area of particular interest was the effect of conducting the cyclic fatigue tests in water, as well as in a relatively

dry environment. In the present paper, Part I, the results from the cyclic fatigue tests are reported and discussed. In Part II [10] the locus of failure of the joints and the mechanisms of environmental attack will be considered. In Part III [11], the results presented in the earlier papers will be used to predict the lifetime of single-overlap joints subjected to cyclic fatigue loading.

2. EXPERIMENTAL

2.1. The Materials

The substrates employed were:

(i) An aluminium alloy (Grade: British Standard 5083), where the main elements, besides aluminium, were 4.0 to 4.9 weight-% of magnesium and 0.4 to 1.0 weight-% of manganese.
(ii) An electrogalvanised (EG) steel which was supplied in sheet form, with a thickness of 1.8 mm, by "ACT Inc." (USA). The galvanised coating on the surfaces of the steel sheet consisted of a zinc coating about 10 μm thick.

The adhesives employed were:

(i) A one-part epoxy-paste adhesive, Grade "XD4600" supplied by Ciba Polymers, UK. This adhesive had been especially developed for bonding aluminium alloys.
(ii) A one-part epoxy-paste, Grade "Terokal 4520-34" supplied by Teroson, Germany. This adhesive is currently being used to bond EG steel parts for the automobile industry.

2.2. Joint Preparation

2.2.1. The Aluminium-alloy/"XD4600" Joints

The aluminium-alloy plate was either 11.0 mm or 12.7 mm in width (*i.e.* "*b*" in Fig. 1) and was machined, using a computer-controlled milling machine, into the tapered-cantilever beams shown in Figure 1. (These values of width, *b*, were more than sufficient to meet the ASTM [12] requirement for plane-strain conditions.) Before bonding, the

substrates were either grit-blasted and solvent degreased (using 1,1,1 trichloroethane) or subjected to a chromic-acid etch [13].

Two pretreated aluminium-alloy beams were then bonded together to form a tapered-double cantilever-beam (TDCB) joint, see Figure 1. A 90 mm length of release-coated aluminium foil was placed at the narrow end of the TDCB joint to act as a starter crack. The thickness of the adhesive layer was 0.4 mm and was controlled by the use of thin wires at the far ends of the TDCB joints. The adhesive was then cured by a two-stage heating process. The joints were initially placed in an oven pre-heated to 145°C for 10 minutes, after which the oven temperature was raised to 190°C. It took about 15 minutes for the oven to reach 190°C, when the heaters were switched off and the oven, and joints, were allowed to cool slowly overnight. A low pressure was applied to the joints during the adhesive curing process.

2.2.2. The EG Steel/"Terokal 4520-34" Joints

The EG steel substrates were only available in relatively thin sheet form, and the sheet was far too thin to be used as beams for a double-cantilever beam (DCB) or TDCB specimens, since even under a relatively small load, gross plastic deformation of the thin arms occurred. (Recall that a requirement for applying the methods of

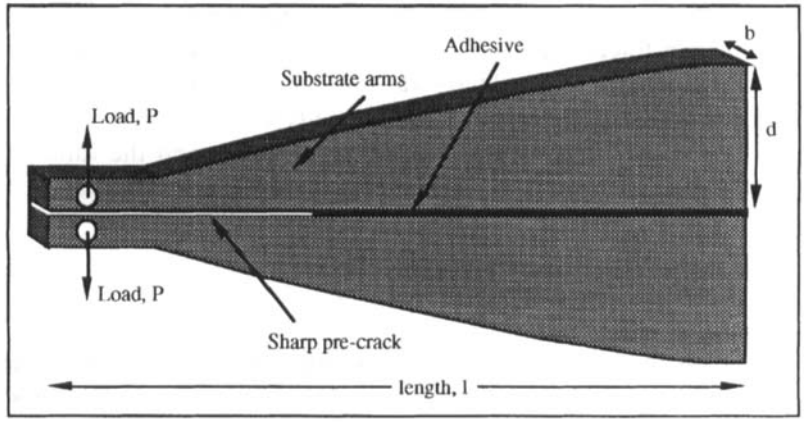

FIGURE 1 The tapered-double cantilever-beam (TDCB) adhesively-bonded joint.

linear-elastic fracture-mechanics (LEFM) to analyse the measured test data is that the arms of the beam must exhibit only elastic deformation). To overcome this problem, previous work by Jethwa *et al.* [14] has developed a "compound" TDCB specimen. In this novel type of specimen the thin EG steel is slotted and bonded into grooved tapered-double cantilever beams of aluminium-alloy, which act as support beams for the thin EG steel strips. Two such "compound" beams are bonded together, so that the EG steel strips are bonded but are supported by the backing tapered-beams of aluminium-alloy. The reader is referred to the previous publication [14] for further details of this "compound" TDCB joint specimen.

The coated surfaces of the EG steel were simply degreased using 1,1,1trichloroethane prior to bonding. The adhesive employed was the "Terokal 4520-34". A 90 mm length of release-coated aluminium foil was placed at the narrow end of the TDCB joint to act as a starter crack. The thickness of the adhesive layer was 0.4 mm and was controlled by the use of thin wire at the far ends of the TDCB joints. The adhesive was cured by heating to 180°C for 30 minutes, after which the oven heaters were switched off and the joints were allowed to cool slowly. A low pressure was applied to the joints during the adhesive curing process.

2.3. Determination of the Adhesive Fracture Energy, G_C

Tests were conducted at a constant rate of displacement of the cross-head of the tensile testing machine in order to ascertain the value of the adhesive fracture energy, G_C. The rate of displacement used for these monotonically-loaded tests was 1.0 mm/min. The tests were conducted at 23 ± 1°C, and the relative humidity was 55%.

LEFM is applicable to the bonded TDCB joints and the value of the adhesive fracture energy, G_C may be calculated *via* the equation:

$$G_C = \frac{P_C^2}{2b} \cdot \frac{dC}{da} \tag{3}$$

where P_C is the fracture load, b is the width of the specimen, C is the compliance ($C = \delta/P$; where δ is the displacement) and a is the crack length. For thin adhesive layers, it has been shown [15] from beam theory for a homogeneous material beam (but not for the "com-

pound" TDCB joint specimen) that:

$$\frac{dC}{da} = \frac{8}{E_S b}\left(\frac{3a^2}{d^3} + \frac{1}{d}\right) \tag{4}$$

where E_S is the modulus of the substrate arms, b is the width of the arms of the specimen and d is the height of the beam at a crack length a. Hence, combining Equations (3) and (4):

$$G_C = \frac{4P_C^2}{E_S b^2}\left(\frac{3a^2}{d^3} + \frac{1}{d}\right) \tag{5}$$

Thus, the value of G_C may be deduced from the measured compliance of the specimen, *via* Equation (3) for any type of specimen, providing the arms of the specimen behave in a linear-elastic manner. Alternatively, assuming the value of dC/da may be accurately described using simple beam-theory, the value of G_C may be also deduced *via* Equation (5).

2.4. Fracture Mechanics Data from the Fatigue Tests

The TDCB test specimen was used to obtain the values of da/dN as a function of the maximum strain-energy release-rate, G_{max}, applied in the fatigue cycle. A sine-wave loading-form was employed at a frequency of 5 Hz. A range of maximum displacements, δ_{max}, were employed in order to cover the complete range of applied fracture energy, G_{max}, values; *i.e.* the range from $G_{max} \cong G_{th}$ up to $G_{max} \cong G_C$. The displacement ratio ($\delta_{ratio} = \delta_{min}/\delta_{max}$) was 0.5.

Displacement, rather than load, control was selected for the fatigue tests since it was found to be easier to detect the lower limit (*i.e.* the threshold value, G_{th}) of G_{max} using the former method of control. This is because with displacement control the value of G_{max} will decrease as the crack propagates through the TDCB specimen, and the crack growth rate, da/dN, will therefore decrease, until it ceases altogether at the value of G_{th}.

It should be noted that G_{max} has been employed, as opposed to ΔG, since during the unloading part of the fatigue cycle the debonded surfaces typically come into contact, resulting in facial interference of the adhesive with itself (if cohesive-in-the-adhesive failure occurs) or with the metal surface (if interfacial failure occurs). This has been observed to lead to the generation of surface debris; which may prevent the crack from fully closing when it is unloaded and hence may give an artificially high value of G_{min}. Thus, it has been suggested [16, 17] that it is better to use G_{max}, instead of ΔG, and this convention has been followed in the present studies. However, the choice of this approach does not significantly affect the general form of the fatigue crack-growth relationships.

For the tests conducted in the "dry" environment, the test temperature was $23 \pm 1°C$ and the relative humidity was $55 \pm 5\%$. For the tests conducted in the "wet" environment, the test temperature was $28 \pm 2°C$ and the joints were immersed and maintained in distilled water for about five minutes before the fatigue tests were started.

The crack length as a function of the number of cycles was determined either (i) by using a travelling microscope, with the side of the TDCB specimen painted white in order that the crack could be seen more clearly, or (ii) by using an automatic data acquisition system [17–19]. This system consisted of using an electrical potential method for measuring the length of the crack. The electrical potential method is an indirect d.c. potential technique and involved the use of a gauge bonded onto the side of the specimen, over the adhesive layer and adjacent substrates. The gauge was a plastic foil with a deposited metal film on its surface. The plastic foil provided both support and insulation from the metallic substrates. A small current of the order of 100 mA was passed through the foil, and when the crack propagated and broke the foil there was a large change in the resistance of the gauge, hence yielding a change in the d.c. potential. The change in potential was relayed from the leads soldered onto the gauge to an amplifier which gave a voltage reading. The signal was then relayed to a Mac Lab data acquisition unit. The Mac Lab was connected to a Macintosh PC. The PC acquired the change in crack length as a function of the time (*i.e.* number of cycles) and a computer program, based on the ASTM Method E647-88 (see below), calculated the rate of crack growth per cycle, da/dN. The PC also acquired the signals of

the maximum load and displacement being applied to the specimen, and, therefore, the corresponding value of G_{max} was deduced. It was found that this method employing the plastic-foil gauge gave extremely accurate and reproducible values of crack length, *a*, *versus* number of fatigue cycles, *N*, for the tests conducted in the "dry" environment, and it was consistently used for these tests. However, for the tests conducted in the "wet" environment a lack of repoducibility was observed. The plastic-foil gauge was covered in a thin layer of polyurethane paint, to protect it from being affected by the water. But it was considered that the errors could have arisen from the gauge preventing water entering the adhesive layer. Whatever the reasons, the values of crack length, *a*, as a function of the number of fatigue cycles, *N*, for the tests conducted in the "wet" environment were found to be more reliable when the method based upon using the travelling microscope was employed. Hence, this was the technique adopted for the "wet" fatigue tests.

The method employed for obtaining values of the crack growth rate per cycle, *da/dN*, was that described as the "incremental polynomial method" in ASTM E647-88 [20]. Several methods have been investigated [17] for deducing the value of *da/dN* associated with a given crack length from the experimental measurements of crack length, *a*, *versus* number of cycles, *N*. The incremental polynomial method was found to be the most accurate, and the one that gave the lowest scatter.

The maximum value of the strain-energy release-rate, G_{max}, applied during a fatigue cycle may be deduced using:

$$\mathbf{G_{max} = \frac{P^2_{max}}{2b} \frac{dC}{da}} \tag{6}$$

where P_{max} is the maximum load applied during the fatigue cycle. Alternatively, assuming the value of *dC/da* may be described using simple beam-theory, see Equation (4), the value of G_{max} may be deduced *via* Equation (7):

$$\mathbf{G_{max} = \frac{4P^2_{max}}{E_s b^2} \left(\frac{3a^2}{d^3} + \frac{1}{d} \right)} \tag{7}$$

3. RESULTS AND DISCUSSION

3.1. Compliance of the TDCB Joints

To validate the experimental techniques, the compliance of the aluminium-alloy TDCB joint was deduced by plotting the compliance, C, *versus* the crack length, a. A typical plot of the compliance, C, *versus* the crack length, a, is shown in Figure 2. The relationship is linear, passing through the origin, and yields a value of dC/da of $2.00 \pm 0.08 \times 10^{-5}$ N^{-1}. The overall experimental value of dC/da was $2.02 \pm 0.16 \times 10^{-5}$ N^{-1}. The theoretical value of dC/da, deduced from Equation (4), for these TDCB joints is 1.80×10^{-5} N^{-1}. Hence, there is good agreement between the experimental and theoretical values, and the value of dC/da is independent of the crack length.

However, it should be noted that the above results were obtained under the "dry" test conditions, either at a constant rate of displacement or under cyclic fatigue loading. When similar studies were conducted under the "wet" cyclic fatigue conditions, then it was found that the experimentally-determined value of the compliance of the aluminium-alloy TDCB joint was often significantly higher than the

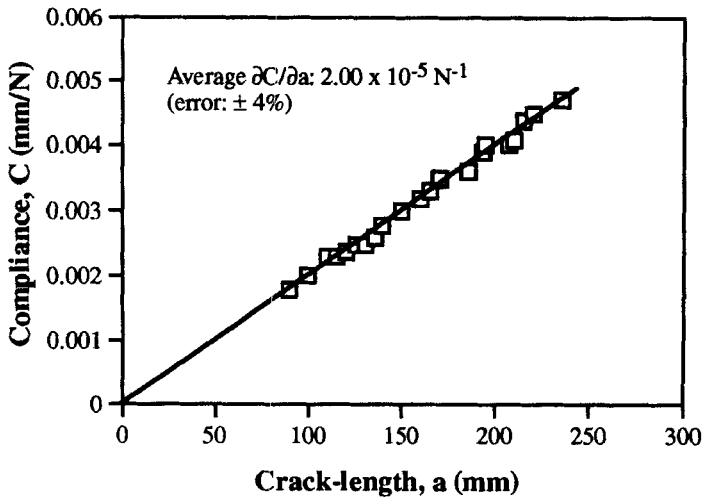

FIGURE 2 Compliance, C, *versus* crack length, a, for an aluminium-alloy/ "XD4600" TDCB joint. (Constant rate of displacement of 1 mm/min; $b = 12.7$ mm).

theoretical value. This was considered to be due to water uptake and plasticisation of the adhesive, particularly in the highly-stressed regions ahead of the crack. These effects would obviously soften the adhesive and could lead to an increase in the compliance of the aluminium-alloy TDCB joint. Therefore, for the fatigue tests undertaken in the "wet" test conditions, Equation (6) was used to deduce the value of G_{max} using the experimentally-deduced value of dC/da.

Finally, as previously discussed [14], Equation (4) is not applicable to the "compound" EG-steel TDCB joints. Thus, for these joints the value of the adhesive fracture energy, G_C and the maximum value of the strain-energy release-rate, G_{max} applied during a fatigue cycle were deduced using Equations (3) and (6), respectively.

3.2. Values of the Adhesive Fracture Energy, G_C

3.2.1. The Aluminimum-alloy/"XD4600" Joints

For the fracture tests conducted at a constant rate of displacement of 1.0 mm/min, the locus of failure was dependent upon the type of surface pretreatment which was used prior to bonding. In the case of the aluminium-alloy substrates which were subjected to a chromic-acid etch, the crack always propagated cohesively through the adhesive layer in a stable manner. The values of G_C determined from either Equations (3) or (5) were in excellent agreement, as indicated by the results discussed above. The measured value of G_C was 3500 ± 125 J/m^2.

In the case of the aluminium-alloy which was grit-blasted and degreased prior to bonding, a mixture of cohesive failure in the adhesive layer and apparently interfacial failure was observed, with the crack again propagating in a stable manner. This locus of failure was reflected in a somewhat lower value of G_C of 3000 ± 65 J/m^2.

Further, for both types of adhesive joint, there was no significant dependence of the value of G_C upon the length of the propagating crack. Hence, no "resistance-curve" (*i.e.* "R-curve") was observed.

3.2.2. The EG Steel/"Terokal 4520-34" Joints

In these tests using the "compound" TDCB joints, the locus of joint failure was always cohesive in the adhesive. The measured value of G_C, from Equation (3), was 740 ± 60 J/m^2.

Again, the crack again propagated in a stable manner and there was no dependence of the value of G_C upon the length of the propagating crack; so no "resistance-curve" (*i.e.*"R-curve") was observed. This observation should be contrasted to that of Spelt *et al.* [21] who have reported an apparent "R-curve" for a similar adhesive. However, it should be noted that these workers have taken the unusual step of defining their experimentally-determined "crack length" as including both the visible main crack and the plastic-yield, or damage, zone ahead of the actual crack. Taking such a non-standard definition will automatically lead to an apparent "R-curve being observed", due to the initiation and growth of the plastic-yield zone during loading the specimen, and very low values of G_C for apparent "crack" initiation will typically be recorded. Also, such a non-standard definition will lead to major problems in accurately and reproducibly defining the initiation of "crack" growth.

3.3. Fatigue Data from the TDCB Joints – "Dry" Environment

3.3.1. The Aluminium-alloy/"XD4600" Joints

The "dry" cyclic fatigue tests were undertaken at a frequency of 5 Hz at a test temperature of $23 \pm 1°C$ and a relative humidity of 55%. As was observed for the constant rate of displacement tests, when a chromic-acid etch pretreatment was used for the substrates prior to bonding, the fatigue tests in the "dry" environment also resulted in the crack propagating cohesively through the adhesive layer in a stable manner.

A graph of the crack growth rate per cycle, da/dN, *versus* the maximum value of the strain-energy release-rate, G_{max}, is shown in Figure 3; as for all such figures, logarithmic axes are employed. The fatigue data, taken together with the value of G_C, reveal a curve which is sigmoidal in shape with three clearly distinguishable regions:

(i) Region I which is a threshold region, and which is associated with very low values of da/dN and G_{max}.
(ii) Region II which is the linear portion.
(iii) Region III where the value of G_{max} starts to approach that of G_C.

In Region I the presence of a threshold value (denoted by G_{th}) below which no significant fatigue crack growth occurs, is clearly visible.

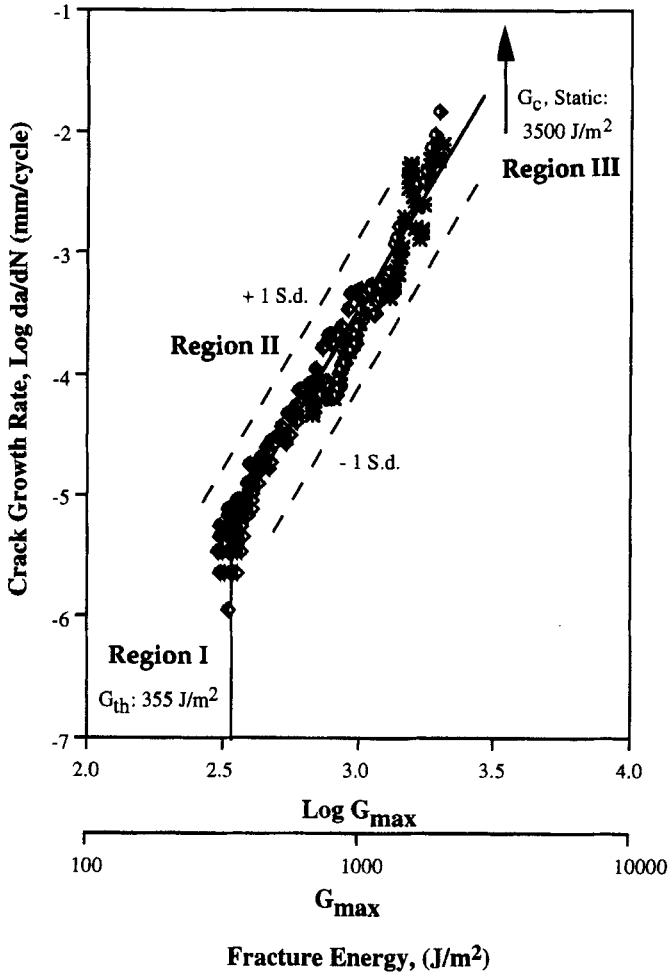

FIGURE 3 Logarithmic crack growth rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the chromic-acid etching pretreatment and were conducted in the "dry" environment of 23°C and 55% RH (For Region II, the limits for $\pm$ 1 standard deviation are shown).

Indeed, the data in this Region I part of the curve show that the values for fatigue crack growth rate, *da/dN*, are approaching 10^{-7} mm/cycle, and this meets the ASTM [20] requirement for the value of *da/dN* to be considered to be negligible. The value of G_{th} is approximately 355

J/m^2. It should be noted that this value of G_{th} is far lower than the adhesive fracture energy, G_C, measured under the monotonic-loading test conditions. Indeed, the value of the threshold value, below which fatigue crack growth is not observed, is approximately 10% of the static value for these "dry" fatigue tests.

In the case of the joints when a grit-blasting/degreasing pretreatment is used, then, as for the constant rate of displacement tests, the locus of joint failure was a mixture of cohesive failure in the adhesive layer and apparently interfacial failure, with the crack again propagating in a stable manner. This locus of failure was reflected, in a somewhat inferior fatigue performance for these joints, compared with those where a chromic-acid etched treatment had been used prior to bonding. This is clearly illustrated in Figure 4. In this Figure the data from Figure 3 for the chromic-acid etched joints are also shown for comparison, but the points have been omitted for clarity. Also, from Figure 4 it is evident that the value of the threshold, G_{th}, for the grit-blasted/degreased aluminium-alloy joints is a little lower, with G_{th} having a value of 250 J/m^2.

3.3.2. The EG Steel/"Terokal 4520-34" Joints

The "dry" cyclic fatigue tests were undertaken at a frequency of 5 Hz at a test temperature of $23 \pm 1°C$ and a relative humidity of 55%. As was observed for the constant rate of displacement tests, the fatigue tests in the "dry" environment revealed that the crack again propagated cohesively through the adhesive layer in a stable manner.

A graph of the crack growth rate per cycle, da/dN, *versus* the maximum value of the strain-energy release-rate, $G_{max.}$ is shown in Figure 5. This shows that a similar relationship exists for this adhesive system as for the alumimum-alloy/"XD4600" joints; compare Figure 5 with Figures 3 and 4. However, although, the "Terokal 4520-34" adhesive has a significantly lower toughness than the "XD4600" adhesive which was used to bond the aluminium-alloy (*i.e.* G_C values of 740 J/m^2 and 3500 J/m^2, respectively) the threshold values, G_{th}, are comparable. This observation is discussed in more detail below.

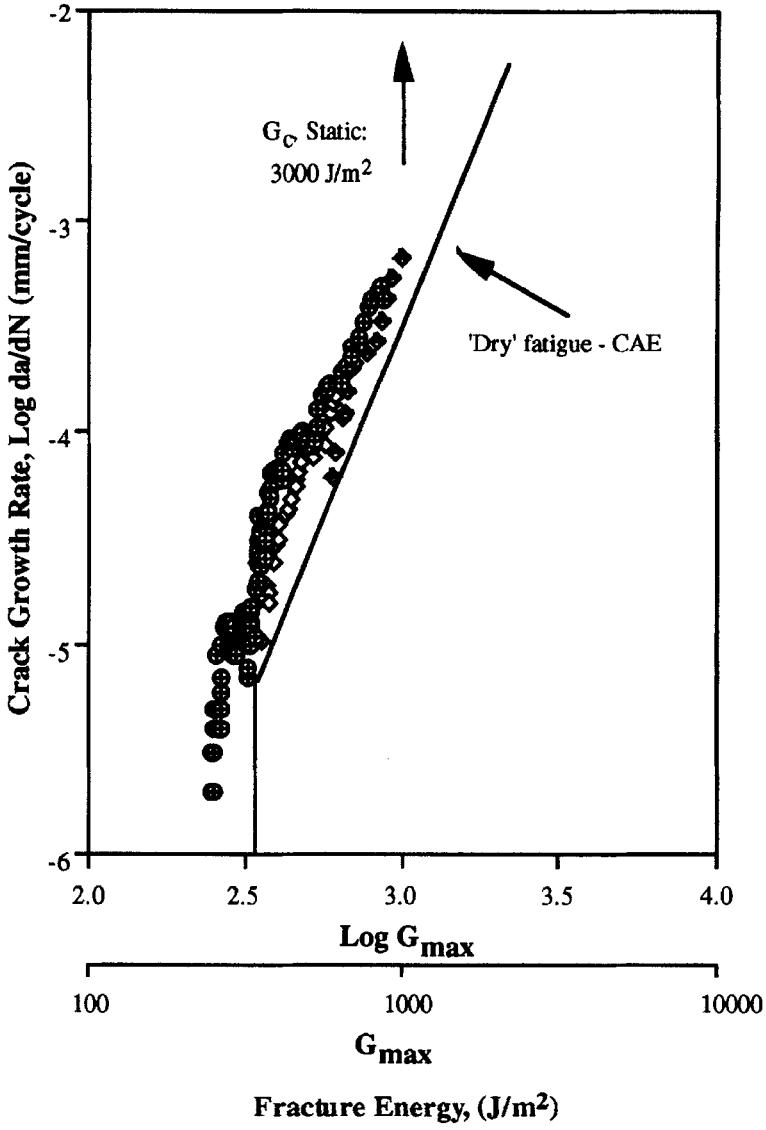

FIGURE 4 Logarithmic crack growth rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the grit-blasting/degreasing pretreatment and were conducted in the "dry" environment of 23°C and 55% RH. (Results from triplicate experiments are shown. The solid line represents the results for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the chromic-acid etching pretreatment and also tested in the "dry " environment. See Fig. 3 for details).

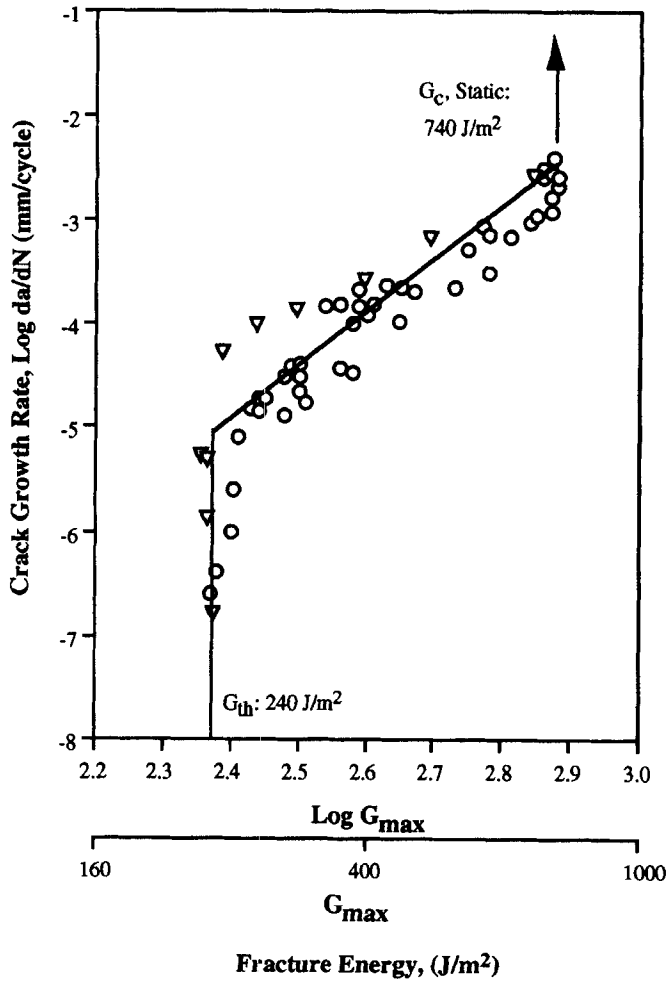

FIGURE 5 Logarithmic crack growth rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the EG steel/"Terokal 4520-34" "compound" TDCB joints and the tests were conducted in the "dry" environment of 23°C and 55% RH. (Results from duplicate experiments are shown).

3.4. Fatigue Data from the TDCB Joints – "Wet" Environment

3.4.1. Introduction

Cyclic fatigue tests were also conducted in the "wet" environment, namely in distilled water at 28 ± 2°C. Again crack growth always occurred in a stable manner.

3.4.2. The Aluminium-alloy/"XD4600" Joints

A graph of the crack growth rate per cycle, da/dN, *versus* the maximum value of the strain-energy release-rate, G_{max}, for the fatigue tests conducted in the "wet" environment is shown in Figure 6. These data are for the joints where the aluminium alloy was subjected to a chromic-acid etching (CAE) pretreatment. As may be seen, the effect of conducting the tests in the aqueous environment is to lower the fatigue resistance of the joints. One aspect of this to lower the value of the threshold, G_{th}, to 200 J/m^2.

The underlying reason for the adverse effect of water on the fatigue resistance of the joints is apparent from a visual assessment of the locus of failure of the joints tested in the "wet" environment. Namely, whilst the joints tested in the "dry" environment failed by cohesive fracture through the adhesive, those tested in the "wet" environment failed by crack growth along the adhesive/substrate interface.

Considering the aluminium-alloy joints when a grit-blasting/degreasing pretreatment was used, then a graph of the crack growth rate per cycle, da/dN, *versus* the maximum value of the strain-energy release-rate, G_{max}, for the fatigue tests conducted in the "wet" environment is shown in Figure 7. The locus of failure for these joints was again visually found to be *via* interfacial crack growth along the adhesive/substrate interface. For comparison, Figure 7 also shows the results from the chromic-acid etched joints which were also tested in the "wet" environment. As expected, the joints prepared using the chromic-acid etch pretreatment possess a markedly superior resistance to attack by ingressing moisture.

3.4.3. The EG Steel/"Terokal 4520-34" Joints

A graph of the crack growth rate per cycle, da/dN, *versus* the maximum value of the strain-energy release-rate, G_{max}, for the fatigue tests conducted in the "wet" environment is shown in Figure 8. As may be seen, the effect of conducting the tests in the aqueous environment is again to lower the fatigue resistance of the joints. For example, the value of the threshold, G_{th}, is lowered to 140 J/m^2. The underlying reason for this adverse effect of water is again the visual observation that, whilst the joints tested in the "dry" environment failed by

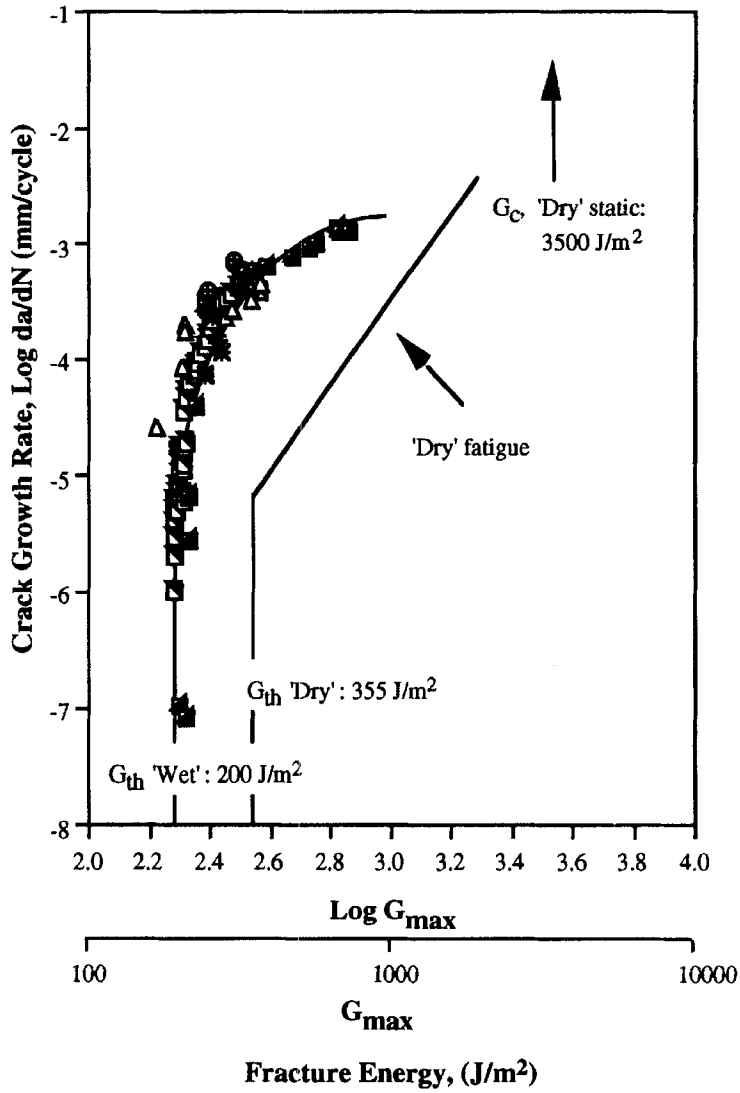

FIGURE 6 Logarithmic crack growh rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the chromic-acid etching pretreatment and were conducted in the "wet" environment of water immersion at 28°C. (Results from six replicate experiments are shown. The solid line represents the results for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the chromic-acid etching pretreatment and tested in the "dry" environment, see Fig. 3 for details).

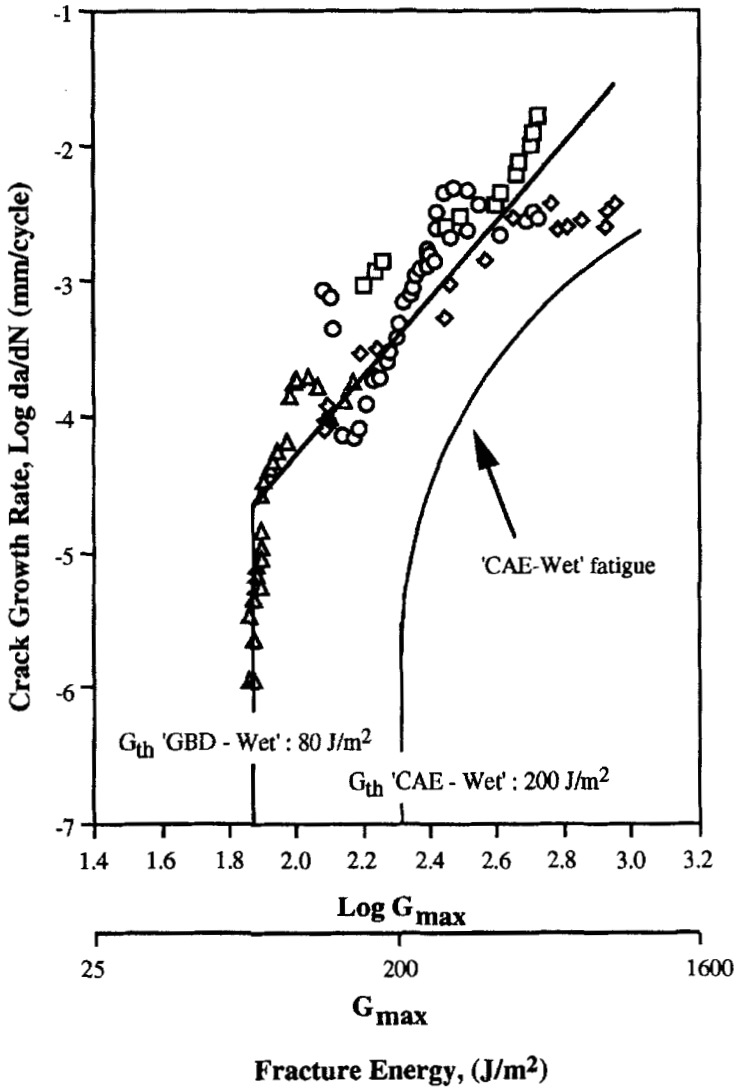

FIGURE 7 Logarithmic crack growth rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the grit-blasting/degreasing pretreatment and were conducted in the "wet" environment of water immersion at 28°C. (Results from four replicate experiments are shown. The solid line represents the results for the aluminium-alloy/"XD4600" TDCB joints which were prepared using the chromic-acid etching pretreatment and tested in the "wet" environment. See Fig. 6 for details).

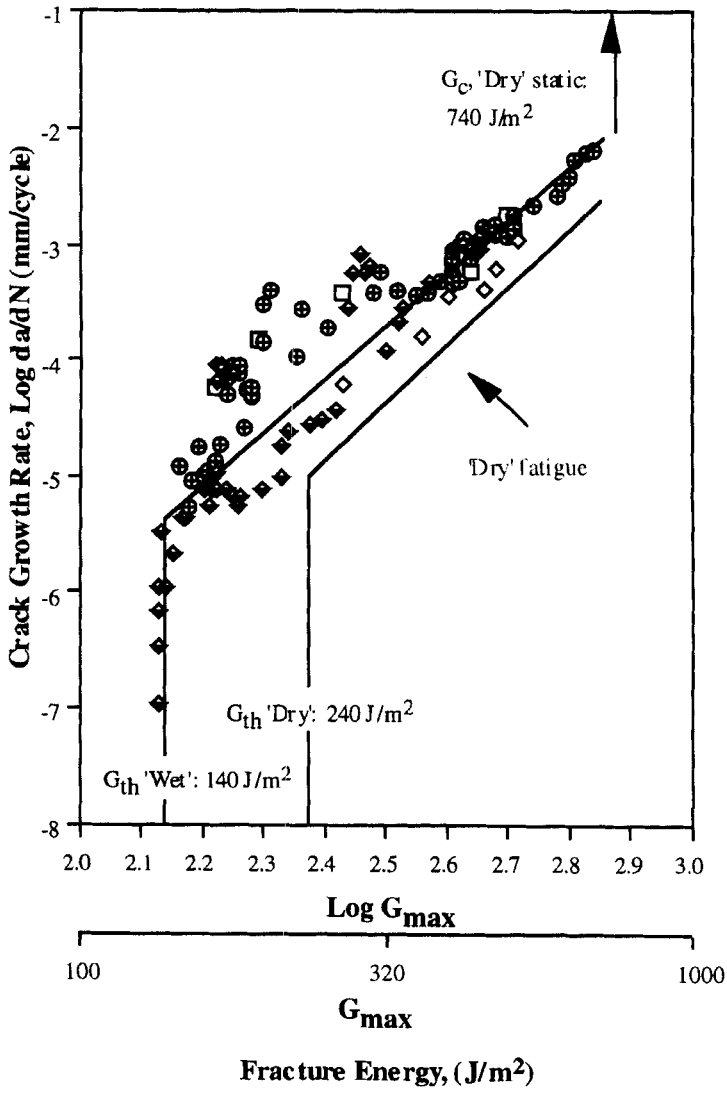

FIGURE 8 Logarithmic crack growth rate per cycle, *da/dN*, *versus* logarithmic, and linear, G_{max} for the EG steel/"Terokal 4520-34" "compound" TDCB joints which were conducted in the "wet" environment of water immersion at 28°C. (Results from four replicate experiments are shown. The solid line represents the results for the EG steel/"Terokal 4520-34" "compound" TDCB joints which were tested in the "dry" environment. See Fig. 5 for details).

cohesive fracture through the adhesive, those tested in the "wet" environment failed by crack growth along, or close to, the adhesive/substrate interface.

3.5. Comparison of Adhesive Systems and Test Environments

The above results are brought together in Table I, which shows the values of the adhesive fracture energy, G_c, the threshold strain-energy release rate, G_{th}, obtained from the cyclic fatigue tests and the appropriate locus of joint failure. They clearly reveal several interesting observations.

Firstly, for the constant rate of displacement (*i.e.* monotonic-loading) tests all the joints gave a locus of joint failure which was cohesive through the adhesive layer, although for the aluminium-alloy/"XD4600" joints where a gritblasting and degreasing treatment was used some apparent interfacial failure was also seen. This mixed locus of failure for these joints explains the somewhat lower value of the adhesive fracture energy, G_c, for the grit-blasted and degreased

TABLE I Comparison of main results

Joint type	*Monotonic tests* $G_C(J/m^2)$	*LoF*	*Fatigue tests* $G_{th}(J/m^2)$	*LoF*
Aluminium-alloy/"XD4600" joints				
"Dry" environment:				
Grit-blast/degreased	3000	Coh./ Interf	250	Coh/ Interf.
Chromic-acid etch	3500	Coh.	355	Coh.
"Wet" environment:				
Grit-blast/degrease	–	–	80	Interf.
Chromic-acid etch	–	–	200	Interf.
EG steel/"Terokal 4520-34" joints				
"Dry" environment:				
Degreased	740	Coh.	240	Coh.
"Wet" environment:				
Degreased	–	–	140	Interf.

Notes:
a. LoF: locus of joint failure.
Coh.: cohesive in the adhesive layer.
Interf.: visually interfacial between the adhesive and substrate.

pretreated joints, compared with the joints which employed a chromic-acid etched aluminium-alloy. The "XD4600" adhesive is clearly basically tougher than the "Terokal 4520-34" adhesive.

Secondly, the "dry" fatigue tests clearly reveal the damaging effect of cyclic loading conditions compared with simply employing a constant rate of displacement to fracture the adhesive joint. Indeed, the value of the threshold strain-energy release-rate, G_{th}, is far lower than the value of the adhesive fracture energy, G_C, which is obtained under monotonic loading. However, the ratio of G_{th}/G_C is not constant for the two adhesives. For the "XD4600" adhesive this ratio is 0.10, whilst for the "Terokal 4520-34" adhesive it is 0.32, thus demonstrating that the basic toughness of the adhesive, as reflected by the values of G_C, is not necessarily reflected in an outstanding fatigue performance of the material. This theme is further illustrated by considering the data shown in Table II. This shows the values of G_C, G_{th} and G_{th}/G_C for a range of adhesives, and in all cases the joints showed a locus of failure which was cohesive in the adhesive. It is clear that a high initial toughness does not necessarily translate into a very high fatigue resistance, at least as judged by the value of G_{th}.

Thirdly, under the "wet" fatigue test conditions the effects of water on the joints are found to be significant. Indeed, the locus of failure changes to an apparent interfacial fracture – at the interface between the adhesive and metallic substrate, or very close to this interface. Accompanying this change in the locus of joint failure, we find that

TABLE II Values of G_C, G_{th}, and G_{th}/G_C for different structural adhesives

Adhesive	$G_C(J/m^2)$	$G_{th}(J/m^2)$	G_{th}/G_C	*Ref*
Hysol "EA9309" (1)	5700	150	0.03	17
Ciba "XD4600" (2)	3500	355	0.10	–
Cyanamid "FM73M" (1)	2930	280	0.10	9
3M "AF-163-2M"(2)	1720	560	0.33	19
Hysol "EA9628 (NW)" (2)	1700	215	0.13	22
Teroson "Terokal 4520-34" (3)	740	240	0.32	–

Notes:
1. (1): carbon-fibre reinforced-plastic substrates
2. (2): aluminium-alloy substrates
3. (3): EG-steel substrates
4. In all cases crack growth was cohesive in the adhesive layer.

the value of G_{th} is greatly decreased, as shown by the data in Table I. The locus of joint failure and the mechanisms of joint attack will be discussed in detail in Part II [10]. However, it is evident that for the aluminium-alloy/"XD4600" joints the values of G_{th} in the "wet" environment are markedly inferior when a grit-blasting and degreasing treatment was used, compared with when a chromic-acid etch treatment was employed.

Finally, it should noted that the fatigue tests at the relatively low maximum displacements (used to obtain the threshold, G_{th}, values) in the "wet" environment typically lasted about one to two weeks. Thus, the fatigue data determined from towards the end of the test were associated with regions of the TDCB joint which had been exposed longer to the aqueous environment than regions from which the earlier test data had been obtained. Now, we have been concerned with the possible interactions between the rate of fatigue crack growth along an aluminium-alloy/"XD4600" TDCB joint (where a chromic-acid etch treatment was used) and water diffusion, and attack on the interfacial regions of the joint, ahead of the advancing fatigue crack. However, we have found that exposing TDCB joints to the aqueous environment, but in an unstressed condition, for up to about six months has no significant effect on the subsequent fatigue curve which was determined in the "wet" environment. Current work is investigating the effect of exposing TDCB joints to the aqueous environment, again in an unstressed condition, for even longer periods of time, prior to then conducting a fatigue test in the "wet" environment.

4. CONCLUSIONS

1. A fracture mechanics approach has been successfully used to examine the cyclic fatigue behaviour of adhesively-bonded joints, which consisted of aluminium-alloy or electro-galvanised (EG) steel substrates bonded using toughened-epoxy structural paste-adhesives. The adhesive systems are typical of those being considered for use, or being used, for bonding load-bearing components in the automobile industry. Fatigue tests were conducted in a relatively "dry" environement of 23°C and 55% r.h. and in a "wet" environment, namely, immersion in distilled water at 28°C.

2. Cyclic fatigue tests conducted in the "dry" environment of 23°C and 55% RH led to joint failure at far lower loads, and far lower values of the maximum strain-energy release rate, G_{max}, applied in a fatigue cycle compared with the value of the adhesive fracture energy, G_C, determined from monotonically-loaded fracture tests.
3. A fatigue theshold value of the strain-energy release rate, G_{th}, could be identified, below which no fatigue crack growth was observed.
4. From studying a range of sturctural adhesives, it is clear that a high initial toughness, as measured by the value of the adhesive fracture energy, G_C, does not necessarily translate into a very high fatigue resistance, at least as judged by the value of G_{th}.
5. Cyclic fatigue tests were also conducted in the "wet" environment, namely, immersion in distilled water at 28°C. These "wet" fatigue tests clearly revealed the significant effect an aggressive, hostile environment may have upon the fatigue performance of an adhesive joint.
6. A major advantage of the "wet" cyclic TDCB fatigue tests is that they may be undertaken and completed in the matter of a few weeks, and do not require the use of unrealistically high temperatures or applied (static) loads in order to accelerate the mechanism of water attack. The use of unrealistically high temperatures or applied (static) loads may actually lead to the joints weakening due to new mechanisms of attack, as opposed to merely accelerating the mechanisms seen in the normal service environment which the joint experiences. Thus, the development and standardisation of "wet" cyclic TDCB fatigue tests may provide the basis for a very effective accelerated-ageing test.
7. The presence of a threshold value of the applied strain-energy release rate, G_{th}, below which no fatigue failure occurs, also has important implications for the design of adhesive joints. Obviously, if the applied loads on the joint are kept below a level corresponding to the value of G_{th}, then joint failure should not be observed, making due allowance, of course, for suitable safety factors. However, it is of importance to investigate whether simply ageing the joints in water (*i.e.* immersing them, under no load, in water) for a relatively long period before undertaking the "wet" fatigue tests would significantly affect the value of G_{th}, and the mechanism of failure. So far, for joints aged in water under no load for up to about six months prior to undertaking the "wet" cyclic fatigue tests, no

effects of the ageing period have been observed. However, longer-term tests are currently underway.

8. In Part II [10] the locus of failure of the joints and the mechanisms of environmental attack will be considered. In Part III [11], the results presented in the earlier papers will be used to predict the lifetime of single-overlap joints subjected to cyclic fatigue loading.

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