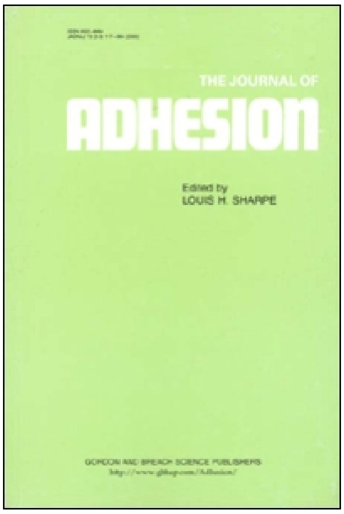

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NOTE

An Examination of Interfacial Failure in Adhesively Bonded Aluminum†

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KEY WORDS Interfacial failure; bond interface structure; transitional regions; environmentally-assisted failure.

INTRODUCTION

It is commonly accepted that the strength of an adhesive bond can be affected by varying the substrate material, substrate surface treatment prior to bonding and the adhesive used in bonding. Although high initial bond strengths can be evaluated and developed relatively easily this area is not usually of major concern to the implementation of bonding technology. Of greater importance for structural bonding applications is the retention of this bond strength with time and the ability of the bonded interface to withstand the presence of defects. The precise nature of the interactions which govern these aspects of behaviour are not currently fully understood.

The initial defect population at a bonded interface may be minimised by ensuring that complete wetting of the substrate by the adhesive occurs. In service, however, environmental interactions may arise which lead to failure at the interface between substrate and adhesive. To help eliminate interfacial failure, aluminium surfaces are generally treated prior to bonding to enhance the long term performance of a bonded joint.

Detailed mechanistic studies of environmentally dominated interfacial failure have provided contradictory evidence for the nature of the interfacial failure mechanism. Physical parameters such as substrate surface morphology have been considered to be important in developing both the initial bond strength and the long term durability.[1,2] Prolonged exposure of phosphoric acid anodised (P.A.A.) aluminium surfaces to humidity results in an oxide to hydroxide conversion. The hydroxide has relatively poor adhesion and it has been postulated that the

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reaction is a precursor to failure with subsequent fracture occurring through the P.A.A. layer.[3] Consequently, corrosion inhibiting surface treatments were found to impart improved joint durability. In contrast, other workers[4] have suggested that the durability is affected not by the stability of the pretreated layer but more by local chemical variations within the pretreated layer.

Although such studies indicate the existence of chemical differences within pretreated surfaces, a correlation between these effects and subsequent failure requires a precise determination of the locus of failure. Such determinations are difficult in view of the limitations of spatial and chemical resolution found in the commonly employed techniques.[5]

While variations in the pretreated layer may be important, a complete understanding must encompass the physical and chemical effects of both the pretreatment and the local adhesive overlayer. Recent studies[6,7] have shown that this interfacial region is not simply a junction of two dissimilar materials but may contain local variations in both structure and chemistry. In general these studies have been conducted using surface treatment procedures that can be applied to small volume components. These treatments generally produce surfaces that have a thick, rough and convoluted surface structure so that a precise understanding of how the interface behaves is difficult to ascertain. For many commercial applications such treatments are not feasible. Recent publications[6,8] have examined the interfacial structure and stability associated with more viable commercial procedures. Although these treatments are of a different nature to those used for aerospace applications they have permitted a detailed examination of the nature of the surface interactions and its significance in determining subsequent bond strength. The research reported here extends earlier observations of interface structure to considering the failure behaviour associated with these interfaces.

EXPERIMENTAL TECHNIQUES

Adhesive joints have been prepared using a commercial Al–Mg–Si alloy and a high-strength, single-part, heat-curing epoxy resin. Prior to bonding the alloy surface was lightly abraded to remove surface asperities the surfaces were then vapour degreased and acid etched with a proprietary solution to remove surface contamination, thereby providing a simple acid etched surface for bonding. In addition, some surfaces were subsequently treated with a proprietary chemical conversion coating to provide a pretreated aluminium surface for subsequent bonding.

Fracture surfaces representing interfacial failure in the substrate surface–adhesive region were produced using an adhesively bonded Al–Mg–Si alloy in a wedge opening load specimen geometry. Interfacial failure was produced by effecting crack growth in a water environment at both 20°C and 60°C.

Examinations of the interfacial region of the bond were conducted using TEM analyses of microtomed sections across the interface, precise details have been given elsewhere.[6] The precise determination of the locus of failure of a bonded joint is difficult to ascertain[5] accurately but is critical to understanding the adhesion between two materials.[9] The techniques commonly used lack either adequate surface spatial or chemical resolution. In view of the practical importance of determining the locus of failure, techniques which incorporate high spatial resolution have been used in the current work.

After failure the fracture surfaces were examined using a high-resolution ISI DS130 SEM and JEOL 2000FX TEM examination of ultra-microtomed cross sections across the interface. To provide edge retention of the fracture surface during specimen preparation the surfaces of several specimens were sputter coated with an impervious layer of gold palladium and mounted in an embedding medium (Figure 1). This prevented damage of the fracture surface edge during specimen preparation and provided a clear indication of the locus of failure since it lay immediately adjacent to the sputter-coated layer. In all cases the features observed in the surface-sputter-coated specimens were also observed in specimens not subjected to sputter coating and embedding.

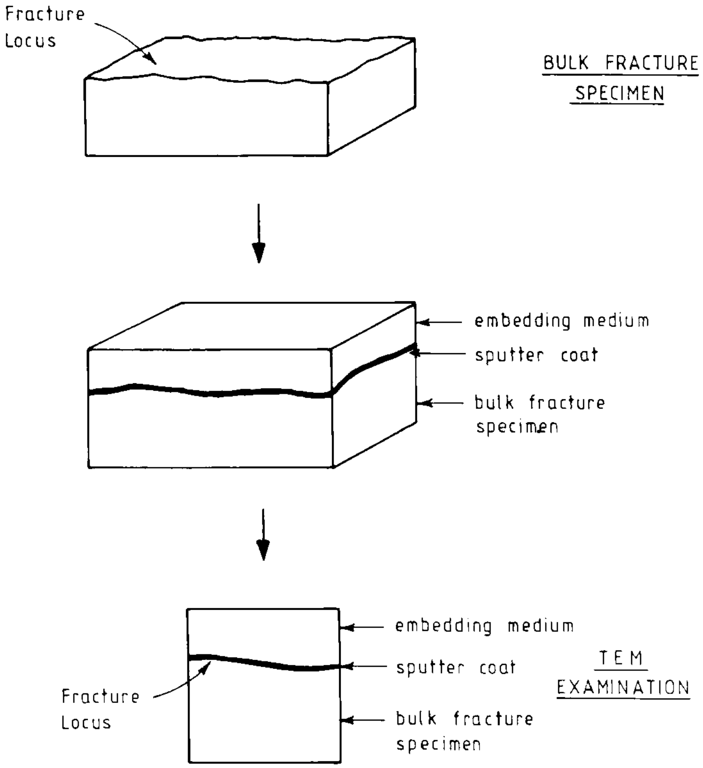

FIGURE 1 Schematic representation of method of examination of locus of failure.

RESULTS AND DISCUSSION

The interfacial region of a bonded aluminium joint has been previously shown to contain local variations in chemistry and structure.[6,8] When viewed as a cross section across the bonded joint these local variations are readily apparent (Figures 2 and 3). Figure 2 shows the transitional region at the interface of a bonded aluminium joint and Figure 3 illustrates that this may contain two transitional regions. Previous examinations of environmentally-induced failure in bonded aluminium have largely ignored the local interfacial variations. Consequently, studies have concentrated on examining either the stability of the substrate surface prior to bonding or the fracture surfaces after joint failure.

SEM examination of interfacial fracture surfaces in the present study failed to distinguish between the surfaces of pretreated-bonded and etched-bonded components when examined immediately behind the propagating crack front. With prolonged exposure of the fracture surface to water, up to 500 h, changes in the simple etched surface were evident; none were observed for the pretreated surface. From previous studies[6] it is known that the presence of surrounding water has little direct influence on the stability of the interfacial region of substrate and adhesive whilst that interfacial region remains intact. A clear discrepancy in interpretation thus arises between these observations and those of the fracture surface after joint failure. As a consequence, the critical events affecting failure of a bonded joint are difficult to interpret clearly. A clearer

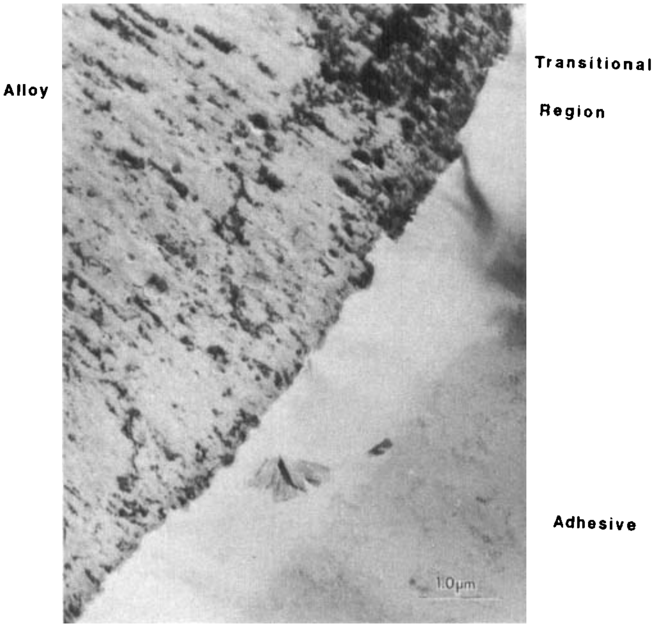

FIGURE 2 Interfacial region showing transition region at aluminium-epoxy interface.

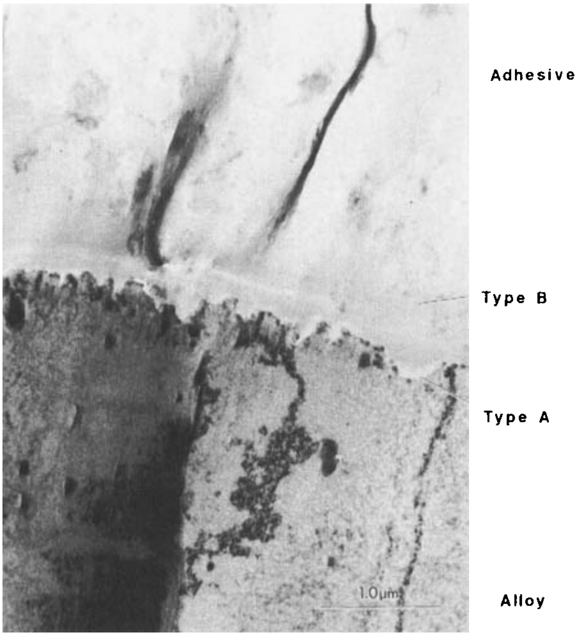

FIGURE 3 Transitional region of bonded interface showing two types of regions commonly observed.

assessment of the failure locus has been made using microtomed sections taken across the fracture surface.

A TEM micrograph of a sectioned interfacial fracture surface of a specimen which had been etched then bonded is shown in Figure 4. The aluminium substrate and sputter-coated layer are evident with the locus of failure immediately adjacent to the sputter-coated layer. At higher magnification it can be clearly seen that failure occurs within the adhesive close to the adhesive–substrate interface (Figure 5). For this surface condition this form of failure was observed to occur at a maximum of approximately 500 Å and a minimum of approximately 10–20 Å from the substrate surface. By examining sections from specimens interrupted at different stages of failure, a thin layer coverage of adhesive on the substrate was observed for locations corresponding to a range of crack growth rates from 10^{-7} mms^{-1} to 10^{-3} mms^{-1}.

A similar examination of environmentally-induced interfacial failure within a joint pretreated prior to bonding revealed identical features (Figure 6). The locus of failure was again observed to be within the local adhesive overlayer for a range of crack growth rates. Due to the penetration of the adhesive into the underlying porous pretreatment a thinner layer coverage of adhesive was generally observed. In this case values ranged from within 200 Å to within 20 Å of the pretreatment surface.

Previous characterisations of the locus of failure of bonded joints have generally been conducted using surface analysis techniques such as XPS, Auger

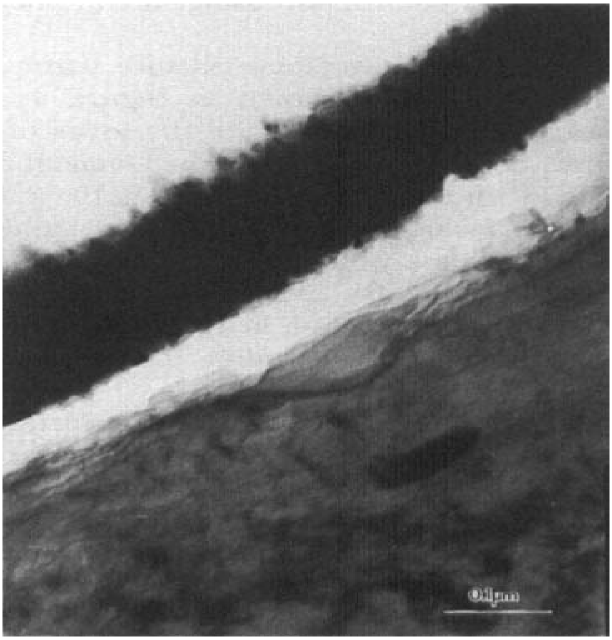

FIGURE 4 TEM micrograph of section through a fracture surface close to an aluminium surface etched prior to bonding.

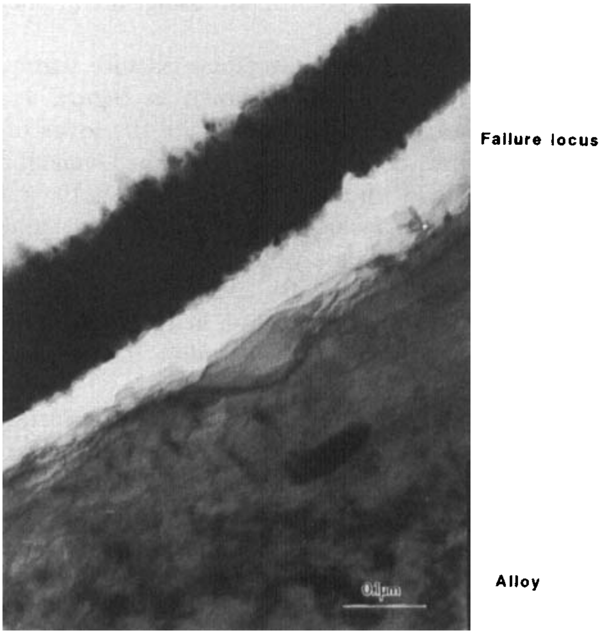

FIGURE 5 Failure of adhesive locally in a joint which had been etched prior to bonding.

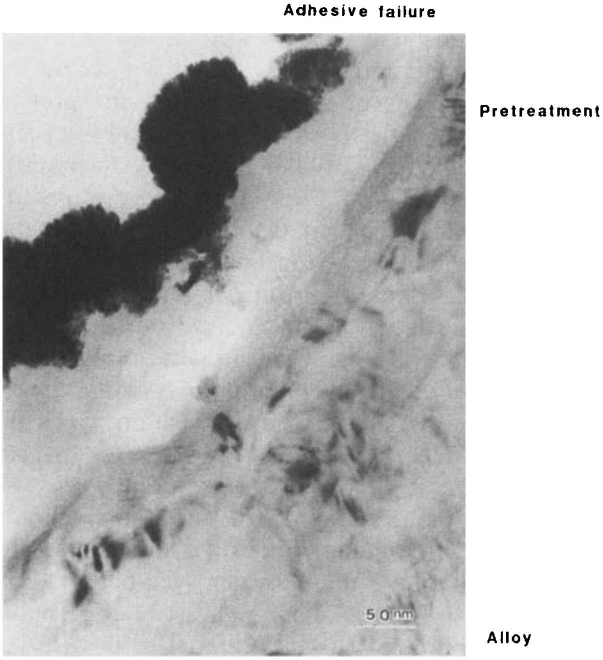

FIGURE 6 Section through a failure associated with an aluminium surface which had been pretreated prior to bonding.

and SIMS. Such techniques usually encompass relatively large area analyses in which initial carbon signals may be ascribed to surface contamination. The present results, however, indicate that a thin layer coverage may arise from failure in the adhesive locally at the interface. In view of the unknown rate at which carbon layers can be sputtered it would be difficult, using surface analysis techniques, to ascribe accurately carbon signals from the current surfaces to either contamination or a thin layer coverage of adhesive. However, the TEM examinations clearly show that the environmentally-induced interfacial failure occurs by failure of the adhesive locally in the interfacial region.

In view of this observation, the existence of transitional layers of adhesive at a bonded interface[6] is significant in determining joint behaviour and failure. Hennemann[10] has recently suggested that differential straining may take place between the various components of the boundary. Although it is not exactly clear what effect this has on subsequent joint failure, clearly the mechanical properties of any localised transitional layers will affect both the local stress distribution and appropriate failure criterion.

Previous examinations of interfacial failure have been interpreted as indicating the locus of failure to be anything from pure cohesive to occurring at the pretreatment–substrate interface. In many cases the environmental failure of bonded aluminium components has been linked to the stability of the aluminium pretreated layer.[1,3,4] However, with such thick (usually > 1 μm) porous surface treatments two points arise. First, it is not clear if the environmental transitions are a pre- or post-failure event and secondly, in such a porous structure the effect of localised transitional regions of adhesive within the pretreated structure is currently unknown. Although the porosity and thickness of such pretreatments make full identification difficult, Brockmann and co-workers[7] have identified transitional layers associated with these complex pretreated surfaces. As a consequence, they have suggested that the chemical constituents of these layers may be important in determining the local corrosion rate of the substructure. In view of the results presented here indicating failure of the transitional region of adhesive locally at the interface, the mechanical properties and stability of the transitional region *per se* may be significant in determining failure (Figures 4–7). In the thick, porous, pretreated layers commonly considered,[3,4] failure of the transitional region of adhesive ahead of a propagating crack tip may result in a local stress redistribution thereby increasing the stresses that are required to be supported by the pretreated structure. At some point these stresses will exceed the local failure stress of the pretreated structure and crack advance will result. Clearly any localised environmental attack of either the transitional or the pretreated layer may enhance the rate at which such failure occurs. However, the current observations of transitional region adhesive failure indicate that not only are the properties of the pretreated layer important in determining failure of a bonded joint but so too are the properties of any transitional regions of adhesive.

In the present case where a pretreatment producing a thin, relatively smooth surface region has been used the failure of these transitional layers primarily governs the rate at which a crack will propagate at a bonded interface. The significance of this result to surface treatments with a more complex structure requires further study to enable a full rationalisation of interface dynamics and chemistry on joint failure to be made.

CONCLUSIONS

The significant structural and chemical variations which can occur at the interface of an epoxy bonded aluminium joint may have a significant influence on determining joint failure. The failure locus of joints visually labelled as occurring at the bond interface was identified to occur within the adhesive adjacent to the metal substrate. This failure results in a thin coating of polymer ≤ 200 Å remaining on the metal surface. The fact that joint failure results in fracture of the polymer in the interfacial regime supports the proposition that it is the mechanical properties and stability of the transitional region of a bond which are important in determining bond strength.

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