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Strength of adhesively bonded joints under mixed axial and shear loading

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ABSTRACT

This study aims at optimising adhesive properties in an aluminium/structural epoxy assembly for different conditions of surface pre-treatment. We consider the mechanical behaviour and failure under proportional, multi-axial loading using an instrumented, Arcan-type test. Values of fracture strength were found to be dispersed (even for a given surface treatment). Typically dispersion was of the order of 15%. This statistical behaviour, also observed with a simple tensile test, seems to be related to the heterogeneous nature of the microstructure of the adhesive bond, which contains voids, as well as mineral particles for reinforcement. A statistical analysis is suggested for use in conjunction with a strength envelope in practical design, for cases when the stress distribution is significantly heterogeneous. It is believed that this approach may be developed in order to understand the well-known scatter of adhesion strength results, and thus contribute to better reliability assessment.

1. Introduction

Joining structural parts with adhesives offers many advantages, compared to traditional techniques making use of mechanical methods of fastening. The aerospace industry has shown a considerable and increasing interest in this technology, which enables dissimilar materials to be joined and generally leads to significant weight saving. Nevertheless, a lack of confidence in ultimate strength limits the use of this technology, due to large dispersion of results, especially after ageing. This statistical nature of strength behaviour can be attributed to various causes, such as defects in the adhesive layer or in surface preparation (either intrinsic or due to inadequate user skill), lack of dimensional precision of the bondline, inhomogeneous ageing, *etc.* Arcan-type tests [1,2] have been employed to obtain fracture envelopes for an adhesive under mixed tension/compression and shear proportional loading, representative of industrial application. The epoxy adhesive studied (Hysol EA9394), when cured, presents a highly heterogeneous microstructure, due to the presence of both reinforcement particles and voids produced during manual mixing of the two components preceding application. The basic idea presented here is to relate dispersion of joint strengths to void and particle sizes and their distributions, an effect which is probably more substantial than dispersion due to any edge effects observed with Arcan test samples.

This preliminary study focuses on the description of protocols and tests, and their interpretation, required to supply design engineers with the data necessary for correct design. In addition, the basics of experimental methodology and physicochemical analysis are considered for quality control of the manufactured adhesive joints. The study points to the limitations of over-simplified mechanical testing and the real need for multi-axial characterisation.

Clearly, a very important consideration in the use of adhesive bonding technology is the nature of the surfaces to be bonded, and their state. Surface preparation is all important. It is well-known that surfaces are often initially contaminated by oil, grease or other contaminants, which must be removed before bonding. This trouble can easily be remedied by suitable polishing, abrasion or washing processes. Nevertheless, for structural loads, such simple treatments are better improved upon.

Metallic substrates are generally covered with oxide films. In many cases, these thin films are only weakly connected to the mother substrate, leading to Weak Boundary Layers (WBL), as promulgated by Bikermann many years ago [3]. As a result, specific treatments have been improved in order to increase the adhesion strength. Because of their use in aerospace equipment, treatment for these materials has been developed more than for others. An outline of a typical recommended process for aluminium and its alloys is as follows [4]. The first step is solvent degreasing, preferably by vapour condensation methods. This is followed by a sodium phosphate/detergent solution wash and then by etching in a chromic acid/sulphuric acid mixture. Finally, the surface is anodised in either a chromic acid or a phosphoric acid bath. Appropriate rinses are applied between each step.

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In the case of bonded joints with aluminium substrates, simple pre-treatment by abrasion and degreasing leads to the lowest failure strength [5].

Provided wetting of the substrate by the adhesive is good (contact angle less than 90°), an increase of adherend roughness should lead to an increase of effective contact area, as predicted long ago by Wenzel [6]. However, it is reported that excess roughness decreases the ability for adhesive penetration, increasing void formation and therefore introducing localised stress concentrations [7,8]. Indeed, a decrease of strength is often found when the adherend surface is too rough. These authors suggested an optimal mean value of arithmetical roughness, R_a, of 3 μm in order to maximise adhesive fracture strength between an aluminium specimen and an epoxy resin. The value of critical roughness depends on many parameters, such as the type of adherend and adhesive, the roughening pre-treatment applied, the joint geometry and the stresses applied. The effect of height and density of peaks on adhesion fracture strength is discussed [9]. An interesting result is that an increase of summit (peak) density is the main cause of variability in the results. An increase in the number of peaks, and a consequent decrease of average distance between peaks enhance the tendency of air bubbles to remain enclosed between the summits, creating stresses' concentrations for the initiation of failure propagation.

Primers and coupling agents are often used to increase the adhesion of structural adhesive bonds, especially those to be used in the presence of water. The pre-treatment of aluminium substrates with silane primers, or coupling agents, prior to bonding can be useful to improve strength. Different surface treatments have been tested with various silane solutions at different pH and per cent proportions. Mohseni and Mirabedini have shown that an aluminium alloy 1050 treated with an amino-silane (at pH 7.5) and with a vinyl-silane (at pH 5) gives rise to an increase of the adhesive surface free energy to about 53 mJ/m^2 and 57 mJ/m^2 [10]. Initially, this alloy showed poor adhesive properties with an apparent surface free energy of *ca.* 23 mJ/m^2 for the untreated surface and *ca.* 33 mJ/m^2 for an acetone degreased surface treatment. Pull-off tests were undertaken subsequent to the different treatments in order to investigate the increase of adhesive strength given with aluminium alloy 1050 after changing concentration of amino- and vinyl-silanes. The highest strength achieved with the amino-silane treatment was observed for a 2% aqueous solution of silane, giving a mean failure stress of 1.8 MPa. For the vinyl-silane treatment, also with a concentration of 2%, a better result was obtained with a mean failure stress of 2.8 MPa under pull-off adhesion testing. This last surface treatment increases the adhesion strength by a factor of 4 by comparison with a simple acetone degreasing surface treatment.

In another study, Deflorian and Rossi compared some surface treatments for corrosion protection of aluminium alloy 5056 [5]. The surfaces were treated with an ethanol solution containing 1.5% of an organo-silane, dipping the surface in the silane bath at pH 7.8 for a few minutes and curing the silane layer for 24 h at room temperature. The adhesion of the coating was measured using a mechanical adhesion pull-off test. This study clearly showed that the silane pre-treatment increased the adhesion strength to a mean of 9 MPa, whereas simple degreasing only reached 4.1 MPa. An increase in the adhesion strength of a factor 2 was thus seen with the silane surface pre-treatment.

Surface treatment clearly affects both strength and *dispersion* of strength of adhesive joints. Adhesive bonds are often loaded in complex stress states and it is therefore of interest to consider the relative effects of tensile and shear stress. The Arcan arrangement is a mechanical test used for the characterisation of adhesive joints under multi-axial loading. It was designed to produce conditions of uniform plane-stress in a specially designed specimen, which is loaded under pure shear/tension or in uniform plane-stress conditions. The Arcan design can be employed to apply proportional, combined shear and tensile loads to butt joint-like test samples, using a tensile testing machine. Such systems have become popular for multi-axial loading since their inception some 30 years ago [1,2], and have been used in many fields such as fracture mechanics [11], and to study polymers and composite materials [12–14].

More complex but reproducible procedures have been developed making use of Arcan fixtures and better-designed test sample geometries. To reduce, or even remove, the stress concentration at the side of the bondline, grooves or beaks are machined at the edge of the adherends [15].

2. Experimental

2.1. Materials and preparation

The substrate material used in this study was an aluminium alloy, 7075 series T, which is widely used in aeronautic and aerospatial applications because of its high mechanical strength and good properties at high temperature [16]. Adherends were machined from the material and are essentially (there is a lip helping attachment) rectanguloid blocks of dimensions $80 \times 10 \times 14$ mm^3, to be bonded on the faces of dimensions 10×80 mm^2. Chamfers of *ca.* 8.5° (with respect to the joint plane) and height 0.15 mm were machined at the corners of the adherend surfaces to be bonded, in order to reduce stress concentration near the edges of the bondline. The value of 8.5° was obtained by finite element analysis to best smooth the edge stress peaks, within the limits set by a flat angled section.

The surface treatments employed in this study were as follows: simple polishing, rubbing with emery paper followed by phosphoric acid anodisation and sandblasting under various conditions with or without subsequent silanisation.

The surfaces were first degreased, then polished (grit size 80), in order to obtain a clean surface for bonding. Various anodising conditions, under both DC and AC, have been tested as pre-treatments for the adhesive bonding of aluminium substrates, as reported in the literature [4]. Anodisation conditions, and resulting structure and thickness of the anodised films, have been shown to affect adhesion and durability of the adhesive joints. In the present case, we opted for anodisation in a phosphoric acid bath for *ca.* 20 min at ambient temperature (*ca.* 23 °C), with a DC voltage of 10 V (see Fig. 1). Before anodisation, surfaces are etched with polishing paper, and then degreased with acetone solution.

Sandblasting of the surfaces to be bonded was undertaken initially using (white) corundum grit of grade F80, having a given grain size in the range of 150–212 μm, and a hardness of

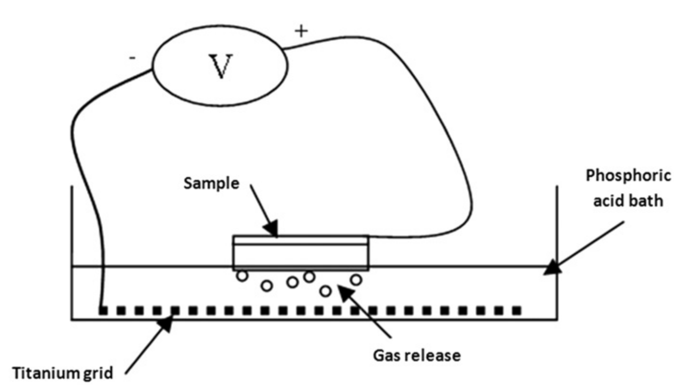

Fig. 1. Schematic representation of set-up for phosphoric acid anodisation (PAA).