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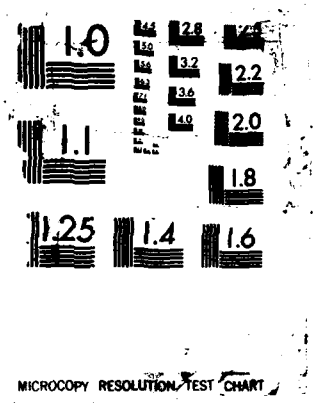
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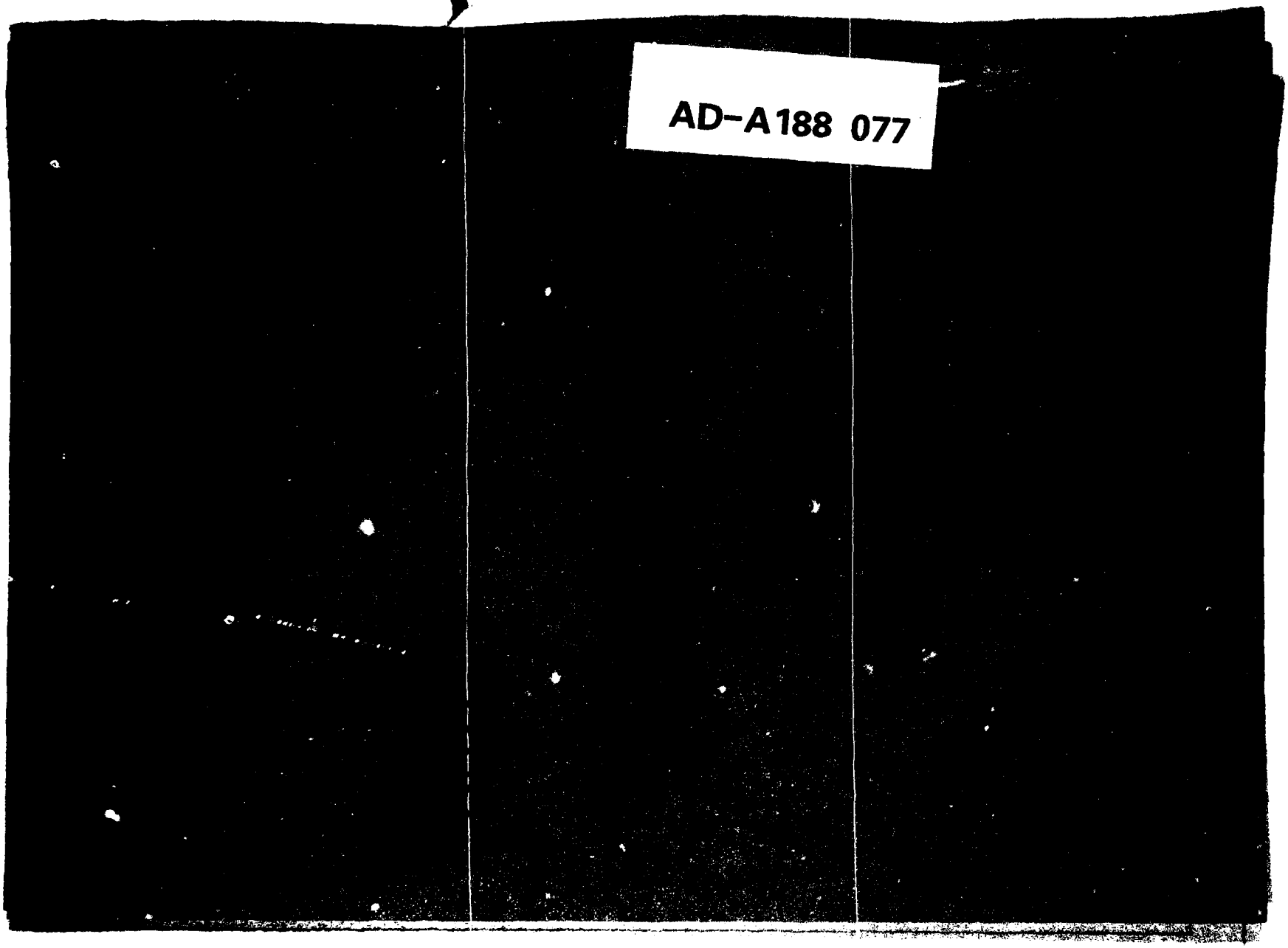

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NON-DESTRUCTIVE METHODS OF CHARACTERISING THE STRENGTH OF ADHESIVE-BONDED JOINTS - A REVIEW

by

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SUMMARY

Progress in the use of adhesive-bonded joints has been hampered by a lack of adequate non-destructive methods to check bond quality. This Report describes briefly how this situation has arisen noting that, whilst many NDT methods give some measure of cohesive strength, it is adhesive strength that is of major concern. For joints with composite adherends it is concluded that it is surface contamination prior to bonding that must be sought, environmental degradation is not a major problem. With metallic adherends, however, the adhesive strength is strongly dependent on the detailed nature of the thin oxide layer and on the way in which this becomes hydrated causing environmental degradation.

Ultrasonic methods are still considered to offer the best prospects and these are discussed in some detail.

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1 INTRODUCTION

Adhesive bonded joints can offer considerable advantages over other forms of joint, particularly in terms of ease of construction and saving of weight, and their use is becoming increasingly widespread. Progress has, however, been hampered by an inability to guarantee that the completed joint is of adequate strength. Because of this, rigorous process control has had to be employed in order to ensure that quality is maintained. In addition it is usual to perform mechanical strength tests on travellers, offcuts or other representative samples. Furthermore, environmental degradation can still give rise to concern. There is therefore an increasing need for more effective methods of Non-Destructive Testing (NDT).

The configuration of adhesive bonds and the materials used can vary widely depending on the particular application. The quality demanded varies equally widely and it is often difficult for the NDT specialist to establish what is required of him. Applications to aerospace structures have, however, tended to set the pace for developments in adhesive bonding, and adhesive bonds in primary aircraft structure are still probably the most critical application. This Report will therefore concentrate on the inspection of these types of joint, although it is hoped that many of the comments will be equally valid for other applications.

Over the years, strenuous efforts have been made to develop non-destructive methods of interrogating the bond and detecting the presence of areas having inadequate strength. As a result a wide range of instruments has been developed but none of them provide a really satisfactory answer. Areas of disbond are generally readily detectable but, as will be shown later, although some correlations of instrument response with bond strength have apparently been demonstrated, such correlations are in fact of limited use. A contributory factor to this lack of success is the rather poor level of communication between the adhesion scientists and the NDT specialists, and the ASE conferences provide an excellent forum within which to try to rectify this. Indeed ASE 85[1] contained some stimulating papers suggesting new approaches. In the author's view, however, there is little advantage to be gained by presenting a review of the various approaches that have been attempted or of the instruments that are currently available. For those that require such information there are already available various useful review papers, such as that by Schliekelman[2]. Although this was published in 1972 it is still largely valid.

Instead, what will be presented is the current RAE view on the nature of the problems, followed by a discussion of some possible routes to a solution. It should, however, be stated that this RAE view was arrived at after a number of discussions with interested parties both in the Aircraft Industry and at Universities. It is recognised that the description of the problems is somewhat simplistic, but it is hoped that it will be sufficient to demonstrate why current NDT methods are inadequate and to indicate what is required in the future.

2 SOME BASIC CONSIDERATIONS

Before considering the requirements for non-destructive methods of evaluating bond strength it is necessary to draw two clear distinctions.

2.1 Adhesive strength and cohesive strength

The first is the distinction between the adhesive strength of the interface (or interfaces) between the adherend and the adhesive layer, and the cohesive strength of the adhesive layer itself. There is sufficient published evidence to enable a reasonable estimate of the latter to be made by measurement of its elastic modulus, density and bond-line thickness. Although the relationship between these parameters and cohesive stength is somewhat empirical some of the existing NDT instruments (such as the Fokker Bond Tester) do in fact measure them, or at least respond to them, in some way. At this stage it should be noted that the response measured is usually dependent on some parameter such as the local mechanical impedance of the structure or a change in resonance of some system coupled to it. Thus any variations in the mechanical properties of the adherend - as could happen when a composite is employed - would complicate the interpretation of such tests.

In contrast, for reasons that will be explained shortly, measurement of the adhesive strength is a great deal more difficult. Indeed a recent authoritative textbook[3] concluded that "The cohesive strength of the adhesive is really the only parameter which can be estimated with any degree of confidence ... ".

Having said this, however, there now seems to be general agreement that cohesive strength is not a matter of primary concern. The fact is that current design stress levels in the bond-line are very low compared with the strength that is normally attainable, and that which is demanded by the mechanical quality control tests. Thus, on the rare occasions when process control fails to guarantee adequate cohesive strength, tests on off-cut coupons or travellers will readily reveal that this has occurred. Furthermore it is unlikely that the cohesive strength could be reduced to an unacceptable level without there being a marked change in the mechanical properties of the layer, which would readily be revealed by NDT. Even those NDT techniques that are primarily intended to look for disbonds will usually respond to other major defects such as gross porosity.

It must be concluded therefore that the most pressing need is to develop improved methods of characterising the interface and hence hopefully to predict the adhesive strength. It is therefore upon this topic that the Report will concentrate.

2.2 The difference between metallic and composite adherends

The second distinction which must be drawn is that between metallic and composite adherends. For the purposes of this Report consideration of composites will be limited to Carbon Fibre Composites (CFC) having epoxy-resin matrices. There is, however, no reason to suppose that the conclusions reached are not equally applicable to aircraft-quality glass fibre composites, although the available evidence is more limited.

In broad terms the joint between a CFC adherend and an adhesive layer is a simple bond between two essentially similar materials. Furthermore, if such a joint is

satisfactory immediately after fabrication then environmental degradation is not a major problem. This statement does, of course, assume that the reduction in strength due to the uptake of moisture (especially that at elevated temperatures) is taken into account in the design stress levels. To this must be added the caveat that there is at present very little known about any additional effects which may be introduced by fatigue loading, but the limited evidence available[4] suggests that fatigue loading at a realistic level has no effect on the residual strength.

In contrast the bond between a metallic adherend and the adhesive layer is an extremely complex one resulting from the procedures necessary to bond two very dissimilar materials. Also environmental degradation is a major problem for such joints. Because of these differences the inspection requirements for metallic and for composite adherends will be examined separately.

3 CFC ADHERENDS

Consider first the situation with a CFC adherend; the interface is not really a plane of weakness and in fact it has been found that a good joint will usually fail in the composite rather than the adhesive (unless a carrier is employed). Furthermore the adhesive layer is no more sensitive to environmental degradation than is the resin matrix of the composite itself and there is no preferential degradation at the interface. The major concern is contamination of the surface of the composite prior to bonding[5] and it is obviously far easier to detect such contamination at that stage. Unfortunately this is rarely possible in a production environment when it is normal practice only to remove the release film at the last moment, with the very objective of minimising the possibility of contamination. Thus the requirement must be to detect any areas having an unacceptable level of contamination in a completed joint. There is at present, however, very little information available on the level at which a given contaminant becomes unacceptable, and RAE and British Aerospace (Warton) hope to mount a programme to establish this for a limited range of contaminants.

Another possible area of concern, although one on which we have limited evidence, could arise if, because of a geometric mismatch, the adhesive were to be cured with minimum or zero compaction pressure. It is possible to visualise a situation in which the adhesive was cured with no contact to one adherend, but in which intimate contact was established when the component cooled to room temperature. In many cases this would only occur at isolated points and would be accompanied by other areas of complete disbond, which would of course be readily detectable. Cases have, however, been reported when quite large areas have suffered from this problem. A typical example is the bonding of a pre-cured curved stringer to a skin. A slight mismatch in the curvatures of the skin and the stringer or distortion of the stringer flanges can result in poor compaction and low bond strength.

The other major problem with CFC adherends concerns the detection of entrapped release film. There would appear to be no fundamental difference here from the detection of similar film in the composite itself. The difficulty of detection is governed by three main factors.

(i) The nature of the film itself: the thin (0.07mm) polyester films are harder to detect than the thicker treated papers.

(ii) The area of film involved: small isolated areas usually stand out, whereas a complete sheet may be harder to detect.

(iii) Whether or not the film has melted: with a melting point of about 170°C this is often a marginal issue.

4 METALLIC ADHERENDS

With metallic adherends the situation is much more complex. In order to obtain a satisfactory bond it is necessary to prepare the metallic surface by etching and/or anodising to produce an oxide layer. Frequently a priming layer of some sort is also necessary in order to protect the oxide and to optimise the bond. The adhesion scientists admit that their understanding of the processes involved is as yet far from complete[6]. It is not even certain how much of the strength is due to chemical bonding and how much to mechanical keying, although a recent paper[7] strongly suggests that microscopic interlocking roughness is a crucial factor. There are in effect three separate interfaces which might need to be characterised:

(i) Metal to oxide.

(ii) Oxide to primer.

(iii) Primer to adhesive.

Of these the oxide-to-primer interface would appear to be most significant, but it should be remembered that failure can also occur within the oxide itself or within the primer. Attention has therefore been concentrated on characterisation of the oxide.

The properties of the oxide that are considered to be of importance are its thickness, its texture and level of porosity, and the degree of hydration. Other factors influencing the strength are the degree of penetration of the primer into the oxide pores and of course the presence of any contaminants.

A further difficulty arises with metallic adherends because, although a substandard surface pre-treatment can have an immediate effect on the as-fabricated strength, its more usual effect is to increase the vulnerability of the interface to environmental degradation. Thus poor pre-treatment will not necessarily be revealed by coupon testing at the time of manufacture.

An improved understanding of the bonding process is, of course, being actively pursued by the various adhesion groups and a wide range of laboratory techniques is being employed in an attempt adequately to characterise the oxide layer and the manner in which it is penetrated by the primer. It is to be hoped that in due course methods will be developed which will enable the surface pre-treatments to be monitored in a production environment. In the meantime, however, it would appear that there is no real substitute for strict process control.

Because of the fact that metallic adherends have of necessity to be subjected to a carefully controlled pre-treatment process it might eventually be possible to introduce

an automated NDT procedure to characterise the resultant oxide. It would, however, be necessary to ensure that there was no risk of the NDT procedure contaminating the oxide or modifying it in some unacceptable way. Even the fact that a requirement for inspection inevitably introduces a time delay between the pre-treatment and bonding stages may well prove unacceptable. With this in mind, it might be suggested that a procedure which could be applied after the oxide is primed would prove more acceptable; but, even if an NDT procedure can be introduced prior to bonding, it should still be remembered that its primary task should be to detect those features of the oxide (or oxide-to-primer interface) that increase the susceptibility of the interface region to environmental degradation. This will clearly not be an easy task.

Because of the above problems the current view appears to be that inspection of the pre-treated adherends is unlikely to be acceptable, and we are forced to consider the possibility of inspecting a completed joint. The difficulty of this task should not be underestimated for, if the subtleties of the interface cannot yet adequately be revealed by inspection prior to bonding, then it is unreasonable to expect subsequent inspection to do so at all readily. Furthermore it is not even the current state of the complex and inaccessible interface that is required to be specified, it is those characteristics of the interface that will govern its future environmental performance.

5 IN-SERVICE INSPECTION

Attention has so far been concentrated upon the potential of non-destructive inspection during and immediately after fabrication. As noted earlier, with CFC adherends it is considered unlikely that there will be a requirement to monitor environmental degradation in service. Disbonds will, of course, require to be found in exactly the same way that delaminations are sought in the CFC. With metallic adherends, however, if the rate of degradation of the bond cannot be sufficiently well controlled by process control or predicted by inspection at the fabrication stage, then it may be necessary to seek some non-destructive means of monitoring this degradation.

What then, are we looking for as evidence that degradation is taking place? Environmental attack by water usually takes place at the adhesive (or primer) - oxide-metal interface. Stress corrosion of the metal substrate is not usually a major mechanism of environmental failure, although it is often a post-failure phenomenon. For aluminium alloys there is clear evidence[7] that the locus of joint failure after environmental attack is through the oxide layer which has been weakened by the ingress of moisture. The weakened oxide is a hydrated form of the original oxide which adheres poorly to the aluminium beneath it. The aim of an NDT technique would be to detect, and possibly to quantify, this oxide transformation. The evidence sought would probably be an increase in oxide thickness and a change in morphology; it is, however, possible that the hydration itself might be detectable by some means.

6 SUMMARY OF PERCEIVED REQUIREMENTS FOR INSPECTION

Before considering possible methods of non-destructive inspection it may be helpful to summarise the conclusions reached in the preceding sections.

6.1 Cohesive strength

The measurement of cohesive strength is not a prime requirement.

6.2 Adhesive strength

This is the area of main concern and there is an increasingly urgent need to be able to characterise the adherend-to-adhesive interface in a completed bond. The features sought are:

(i) Composite adherends

Surface contamination.

Lack of compaction pressure.

(ii) Metallic adherends

The nature of the oxide layer.

Changes in the oxide layer caused by environmental exposure.

7 THE FOKKER BOND TESTER

As noted in the Introduction it is not considered very productive to attempt to review those commercial instruments that are currently available. Some brief comments on the Fokker Bond Tester are, however, considered worthwhile, not only because of its widespread use over many years, but also because various apparent correlations have been demonstrated between its response and cohesive lap shear strength.

The instrument is fully described in Part 2 of Ref 2, but it essentially consists of a piezoelectric transducer which is capable of being driven at different frequencies and which is coupled to the component by a thin layer of liquid or gel. The resonance characteristics of the bonded joint together with those of the probe are analysed by sweeping the driving frequency through a range in which certain modes of vibration are excited. The response is shown on two displays. The A-scale displays the frequency response and the B-scale gives a measure of resonant amplitude.

A recent paper by Guyott et al[8] has examined the possible resonances in the probe crystal and modelled the probe-specimen interaction using a receptance analysis. This showed that there were two main modes of vibration. The first mode of vibration tended towards that of a simple mass-spring-mass system, with the mass of the bottom adherend coupled by a spring (the adhesive) to the mass of the probe plus the top adherend. At lower adhesive stiffnesses a second mode of vibration occurred in which the probe and top adherend no longer moved as a rigid body, and the bottom adherend became progressively decoupled from the system.

As shown in Fig 1, the analysis predicts that the resonant frequencies corresponding to these two modes are essentially functions of the specific adhesive stiffness, which they defined as the ratio of apparent adhesive modulus, E_a, to adhesive thickness, t_a. The effect of adhesive density was shown to be small. The figure also shows how the predicted behaviour was confirmed by experimental measurements. These were made on a range of single lap joints with 1.6 mm thick adherends and an adhesive

thickness range of 0.05 to 0.75 mm. Two different adhesives were used, representing the extremes of modulus considered likely to be encountered in practice.

These results are quite encouraging but they do emphasise that it is not possible to distinguish changes in adhesive thickness from variations in modulus. Now both these parameters can affect the cohesive strength and it might be necessary to distinguish between them. For example, as Guyott et al point out, both very thin or very thick bond-lines can result in a reduction in strength and it would certainly be possible to have two joints having the same specific stiffness but exhibiting very different strengths. On the other hand it is also possible to envisage a processing defect that would result in a porous bond of excess thickness; this would cause a decrease in E_a and an increase in t_a, both of which decrease the specific stiffness. It is, however, not possible to generalise and it must be concluded that, while the Fokker Bond Test may be able to reveal changes in cohesive properties, it is essential to understand the changes in parameter that are being sought if ambiguity is to be avoided.

8 ULTRASONIC METHODS

8.1 Normal incidence

In all the numerous attempts to develop a non-destructive means of assessing the quality of an assembled joint, the vast majority have used some form of ultrasonic interrogation, and ultrasonic methods still appear to offer the best prospect of providing a practical method in the near future. Most of these attempts have used a conventional pulse-echo approach in which the direction of the ultrasonic wave is normal to the plane of the adhesive layer. Electronic gating or sampling is used to isolate echoes from specific interfaces or sets of echoes from more than one interface. In order to identify the small changes in these echoes, which are very hard to distinguish in the conventional time-domain presentation, it has usually been necessary to process the signals and to present the data in the form of frequency spectra (and occasionally even cepstra).

Interpretation of the resultant spectra is, however, very difficult and various attempts have been made to model the propagation of ultrasonic waves in multilayer laminates; there has also been a good deal of supporting experimental work. Most of the models have, however, been highly idealised and have really so far only been of assistance in characterising the cohesive properties. Two convenient reviews of work in this area are available[9,10], and it is clear that strenuous efforts have been made to refine the techniques and to extract the maximum amount of information from them. So far, however, the only real success has been in obtaining correlation with cohesive properties (see, for example Alers et al[11]). Indeed, in his revue[10] Curtis included the statement that "The technology required to examine the associated time and frequency domain exists but the 'Holy Grail' of adhesion strength still resolutely defies non-destructive evaluation by pulse-echo means".

Essentially there are four parameters which primarily affect the ultrasonic response in a simple pulse-echo system:

(i) The transit time of the ultrasonic pulse through each of the two adherends.

(ii) The transit time of the ultrasonic pulse through the adhesive layer.

(iii) The reflection (and transmission) coefficients at the adhesive-to-adherend interfaces.

(iv) The attenuation experienced by the pulse as it is transmitted through the adhesive layer. (For thin bond lines this is probably a secondary factor.)

Now the transit time in the adherends is effectively constant in metallic adherends of uniform thickness, but can vary significantly from point to point in composite adherends and this might tend to mask changes in the other three parameters. It should also be noted that the moisture uptake which is inevitable during the service life of composite adherends will also affect the transit time, but if it is not necessary to monitor environmental degradation in such joints then this may not be important. Furthermore the attenuation, which is negligible in metallic adherends, may prove significant if composite adherends are employed.

The transit time through the adhesive layer is dependent on the velocity in that medium and on the thickness of the layer and, as was found for the Fokker Bond Tester, it may be difficult to separate the two parameters and to obtain a corrrelation with cohesive strength. It has, however, been demonstrated[11] that, if the bond line thickness is maintained constant and the cohesive properties of a two-part paste adhesive varied by using different proportions, then an empirical linear relationship exists between the velocity of sound and the lap shear strength. It should be noted that the velocity, and hence the transit time, will change if the adhesive layer takes up moisture and it will be shown in the next section that this may complicate the interpretation of tests aimed at revealing environmental degradation at the interface.

The reflection and transmission coefficients on the other hand are primarily governed by the relative values of the acoustic impedance (the product of wave velocity and density) on either side of an interface between two materials. They will, however, also be affected by the nature of that interface and a number of questions remain to be answered.

(i) Is the complex nature of real interfaces sufficiently well understood to permit a model to be constructed?

(ii) Is a mathematical solution to the model available?

(iii) Do the factors affecting adhesive strength play a part in the model that is sufficiently significant for them to be quantified?

(iv) Are there practical variations in real joints which make the model unrepresentative or inapplicable?

Consideration will now be given to each of these questions and, to avoid confusion, the situation for metallic and for composite materials will be examined separately.

8.1.1 Metallic adherends

The nature of the oxide layers produced by the various pre-treatment processes, and of the resultant interfaces, has been investigated quite intensively for many years (see, for example, Refs 7 and 13) but there appears to be little information on their mechanical properties such as elastic modulus and density which would be required by the models.

The oxide thickness itself can vary very widely depending on the process used, as is demonstrated by the following table.

Process	Average oxide thickness nm
Chromic-sulphuric pickle	40
Phosphoric acid anodise	400
Chromic acid anodise	2000
Wavelength of a 10 MHz compression wave in aluminium	600 µm (600000 nm)

It should also be noted that even the thickest layer is some two orders of magnitude less than the wavelength of the interrogating ultrasound. This must clearly be recognised when considering the validity of the models.

The presence of a thin intermediate layer, such as an oxide film, will effectively provide a frequency dependent modulation of the fundamental reflection coefficient. The magnitude of this effect is dependent upon both the thickness of the layer and its acoustic impedance. The better the acoustic match between the oxide and the aluminium adherend the less will be the degree of modulation. No data appears to be available on the acoustic impedance of a representative oxide but, since text-book values for an unspecified form of aluminium oxide are nearly twice that of aluminium, one is led to wonder whether a porous oxide has an impedance which is similar to that of the parent sheet. If that were the case then it would not be detectable.

Fig 2, which is taken from Kwakernaak et al[13], shows schematically the pore structure which they consider is produced by the European chromic acid anodising process; the average pore wall thickness is estimated to be about 15 nm. With such a narrow, deep pore structure the extent to which the adhesive or primer can penetrate the pores must be debatable, yet this degree of penetration will clearly affect both adhesive strength and the way in which the interface should be modelled. It is likely that there are similar uncertainties associated with the other oxide structures. There is in fact still considerable uncertainty regarding oxide morphology, and Kwakernaak noted that the model that he had derived was quite different from that proposed by Venables[7] (Fig 3). Since Venables placed considerable emphasis on the role played by mechanical interlocking it is clearly essential that conflicts of this nature be resolved.

Thus at this stage, modelling of the interface characteristics would appear to be somewhat arbitrary. Most of the models used so far have used either an extra thin intermediate layer to represent the oxide[11] or a so-called 'slack' boundary[14] which can stretch and behave elastically under stress. Both of these approaches appear useful and it should certainly be possible to predict the difference in ultrasonic response expected from significantly different pre-treatment processes. The fact that there is a marked difference in response has been demonstrated by Mahoon[15], who used a frequency domain display to distinguish between the responses of an etched surface and one with additional chromic acid anodising.

Mathematical solutions are available. Most of these have been based on the classical formulae of Brekhovshikh[16], but recent work by Challis et al[14] using the slack boundary model used a Laplace transform approach to give the response function of the adhesive layer together with both interfaces. The latter approach is analytically tractable only for a restricted range of simple input functions, and a discrete time equivalent has to be used to encompass input waveforms of arbitrary shape. Only limited experimental data have so far been presented.

So far then the picture seems comparatively hopeful, but now we must turn to question (iii). Do the factors affecting adhesive strength play a part in the model that is sufficiently significant for them to be quantified? To this, at present, there would seem to be no easy answer. As discussed in section 4, what is primarily sought is a means of revealing those features of the oxide, or of the interface, that increase its susceptibility to environmental degradation. If those features have not yet been properly identified then it is not possible to examine whether they could be introduced into the model in a meaningful fashion. Venables[7] has presented evidence which suggests that the most important feature of the oxide, with regard to the initial quality of interface adhesion, is that it should have a morphology such that it can interlock mechanically with the adhesive. Modelling this feature may not be easy but must certainly be regarded as feasible.

The long-term durability of such bonds is, however, largely determined by the environmental stability of the oxide. For aluminium, moisture ingress at the bond line causes the oxide to convert to a hydroxide which adheres only poorly to the aluminium substrate. Venables went on to demonstrate that significant improvements in environmental durability could be achieved using an extremely simple treatment whereby monolayer films of certain organic acid molecules are used to protect the oxides from the effect of moisture. Furthermore he showed that the oxides formed by the phosphoric acid anodising process, which are normally stabilised to some extent against moisture by absorbed phosphate, can be stabilised even more effectively by an amino phosphonic acid treatment. The ability of a model adequately to represent such a monolayer must be highly debatable. It may perhaps be necessary to approach the problem from the other direction by gathering experimental data from specimens having carefully controlled variations in pre-treatment and, seeing if a sufficient difference in response is obtained. Such an approach would initially have to be empirical, but it might then be possible to modify the model in order to conform to the observed data. This would be

very valuable since it would permit an assessment of the practical utility of the empirical correlation, ie whether or not the significant features could in practice be masked by changes in other parameters.

The situation is more hopeful, however, when we consider the possibility of monitoring in-service degradation. There is a fair amount of information available on the mechanism of hydration of the oxide. The hydroxide is much thicker than the original oxide and there is a marked change in morphology so it should readily be capable of being modelled. It should, however, be remembered that the velocity, attenuation and acoustic impedance of the adhesive layer itself will also be affected by moisture uptake. Now, moisture uptake in the adhesive may not be too serious, it is the weakening of the interface that must be detected and most of the past investigations have not been able to separate the two parameters. Improved modelling may well be able to rectify this.

The final question is whether real bonded joints depart sufficiently from the idealised joint of the model to make the analysis invalid. With metal adherends the two main problems would seem to be the possible variation in thickness from point to point and, arising from this, a lack of parallelism of the adherend surfaces. It should be noted, for example, that Alers et al[11] included spacers to ensure parallelism, and Challis et al[14] stated that any slight angulation between the faces would result in severe signal distortion due to wave diffraction phenomena. Practical aspects of this nature must clearly not be overlooked since they may severely limit the amount of information that is obtainable.

8.1.2 Composite adherends

If contamination is indeed the major problem in bonds with composite adherends then it is possible that it might be revealed as a simple change in reflection coefficient and that no sophisticated modelling may be necessary. This comment is based on some preliminary results obtained by Curtis[12] who used a high frequency time domain presentation to separate the echoes from the front and back interfaces in a simple lap joint. He introduced various degrees of contamination by Frekote 33 (a release agent) and was able to demonstrate an empirical relationship between the relative amplitude of the echoes and the lap shear strength. It is not clear, however, how representative was the degree of contamination. It also seems likely that the texture of the composite surface will have a strong influence on any such relationship. The contamination left by an impregnated glass release cloth, for example, is likely to occur in a series of peaks and troughs.

If the above approach does not prove satisfactory then it may be possible to model the weakened interface as a very thin intermediate layer of a slack boundary. It should, however, be recognised that the inherent variability in the properties of the composite adherends will make this task a great deal more difficult.

There is little evidence so far on the effect of low compaction pressure. The majority of adhesives are able to flow quite freely and this enables a fair degree of mismatch to be accommodated. If it cannot be accommodated then, apart from an actual

disbond, the most likely effect is to produce a degree of porosity which will alter both the acoustic impedance and the attenuation characteristics of the adhesive layer. The thickness of the layer will also, of course, be excessive. It is hard to visualise a situation in which the properties of the adhesive layer are satisfactory but in which poor compaction has produced a weak interface. If this does occur then, from a modelling point of view, it may not be too dissimilar to the situation with contamination at the interface.

8.2 Surface and interface waves

As was noted earlier, the thickness dimension of the interface layers is in general very much less than the wavelength of the ultrasound and it is acknowledged that normal compression waves of the type considered in section 8.1 are insensitive to the existence at the interface of a thin liquid-filled layer which exhibits no shear resistance. Rokhlin[17] showed how, if this layer is thin enough, there can be complete transmission of compression waves, despite the fact that the interface has no shear resistance and suggested that it would be more meaningful to interrogate the bond with interface waves. Such waves produce shear stresses at the interface and propagate along the interface; they are therefore sensitive to variations of adhesive quality. He went on to propose the model shown in Fig 4, which assumes that the viscoelastic properties of the adhesive are not uniform over its thickness and that a reduction in the elastic modulus may occur at the adhesive-adherend interface. Adhesive (interfacial) failure is identified with the failure of a weak boundary layer (WBL).

As shown in the figure ρ and μ are the density and shear modulus of the adherend and suffices 0 and w identify similar parameters in the adhesive layer and weak bond line respectively. The thickness of the adhesive layer is $2h_0$ and the thickness of each WBL is h_w .

He introduced the concept of an effective shear modulus μ_{eff} which is calculated on the basis of the measured velocity and attenuation of interface waves. This is related to μ , the shear modulus of the adherends, by a rather complicated expression involving h_0 , h_w , ρ , ρ_0 , ρ_w , the frequency f and the following three velocities,

V_t the transverse (shear) wave velocity in the adherend

V_l the longitudinal wave velocity in the adherend

V_i the interface wave velocity.

Thus if V_i is measured experimentally then μ_{eff} may be calculated.

The ratio of μ_{eff} to the shear modulus μ_0 of the adhesive layer is given by

$$\frac{\mu_{eff}}{\mu_0} = \frac{1 + \frac{h_w}{h_0}}{1 + \frac{h_w \mu_0}{h_0 \mu_w}} \qquad (1)$$

and this can be used as a criterion of the bond strength. If μ_0 is measured on a reference specimen then equation (1) can be used to estimate the properties of the WBL.

When contact between the adhesive and the adherend is not ideal the interface velocity decreases and the effective shear modulus μ_{eff} will be smaller than μ_0 the actual shear modulus of the adhesive layer. In the limit, where there is an absence of shear bonding between adhesive and adherend, the velocity of the interface wave will approach that of the Rayleigh wave and the calculated value of μ_{eff} will be zero.

He was able to show experimentally[18] (see Fig 5a) that if a pre-treated metal adherend is contaminated with an infinitely thin lubricant film prior to bonding then there is a good linear relationship between the resultant normalised shear strength and the normalised effective modulus. No information was given, however, on the nature of the surface of the pre-treated adherend. He also showed (Fig 5b) that the shear strength can be correlated with a transmission loss factor which embraces attenuation and scattering phenomena. Some supporting evidence for this was provided by Claus and Kline[19] who bonded borosilicate crown glass to Pyrex using an anaerobic cement. A range of surface finishes was introduced into the crown glass specimens prior to bonding by polishing the surfaces with different grades of carborundum optical abrasive. Measurement of the attenuation of interface (Stoneley) waves readily revealed the presence of the different surface finishes. They suggested that similar methods might be used to assess the effects of chemical contamination.

Some complementary evidence is provided in a recent review by Pilarski[20]; he showed that the quality of a duralumin to epoxy resin bond could be related to the phase velocity of various wave modes, a higher phase velocity corresponding to a greater degree of adhesion.

At present therefore it can only be stated that, although interface waves would be more difficult to generate and detect, they may be able to provide more information about the state of the oxide-to-primer interface for a fully cured adhesive.

9 OTHER NDT METHODS

As was stated in the Introduction, there are many commercial instruments available, but most of these rely on the generation of some form of mechanical vibration and are not sensitive to those parameters which this Report suggests are important. A useful review of the essential principles involved is given by Curtis[10]. He also discusses the possibility of using the acoustic emission technique which is not truly non-destructive but might be used in conjunction with a proof test. In this technique a mechanical load is applied and sensitive transducers used to detect the small stress waves that are generated by individual failure events. The hope is that the acoustic emission characteristics measured on a component at some modest load level could be used to predict the failure load on that component. Limited success has been achieved on simple specimens bonded with certain adhesives and exhibiting a mixture of cohesive and adhesive failure. However, other adhesives, particularly those exhibiting a high peel strength, do not emit at all until failure is imminent. There appears to have been little recent activity on this topic which may well indicate that it is considered of limited value.

Overall there seems to have been somewhat of a dearth of new ideas. A new approach to the characterisation of cure in epoxy resins has, however, been described by Affrossman and Pethrick[21]. In their method, termed Thermally Stimulated Discharge (TSD), electrostatic charges are induced into the resin at a temperature just below its T_g and are trapped when the sample is cooled. When the sample is slowly re-heated the trapped charges are released and can be measured as a current flow. A trace of the current flowing in the sample against temperature contains features which reflect the electrical characteristics of the resin. They have demonstrated the ability of TSD to reveal the degree of post cure and to monitor the uptake of moisture but the full potential of the method has yet to be explored.

10 CONCLUSIONS

(1) Although improved prediction of cohesive strength could be useful it is adhesive rather than cohesive strength which is the matter of primary concern.

(2) With carbon fibre composite adherends the main concern is contamination of the surface prior to bonding. If a joint is satisfactory immediately after fabrication then environmental degradation is unlikely to be a major problem.

(3) With metallic adherends it is the detailed nature of the oxide produced by the etching and/or anodising process that is of prime concern since this governs the adhesive strengths.

(4) With metallic adherends environmental degradation occurs by hydration of the oxide; the mechanism by means of which certain pre-treatments can delay the hydration process is not entirely clear but appears to be associated with the provision of a protective monolayer film.

(5) Characterisation of the morphology of the oxide may permit the as-manufactured adhesive strength to be predicted.

(6) In order to ensure long-term environmental durability it may be necessary to detect the presence of the protective monolayer.

(7) Characterisation of the oxide or detection of the protective monolayer would probably be much simpler prior to the assembly of the joint, but this may not be acceptable in practice.

(8) Ultrasonic interrogation currently appears to offer the best prospects for in-service inspection in the near future, although there is as yet no adequately developed method.

(9) There is still no method capable of predicting susceptibility to environmental degradation.

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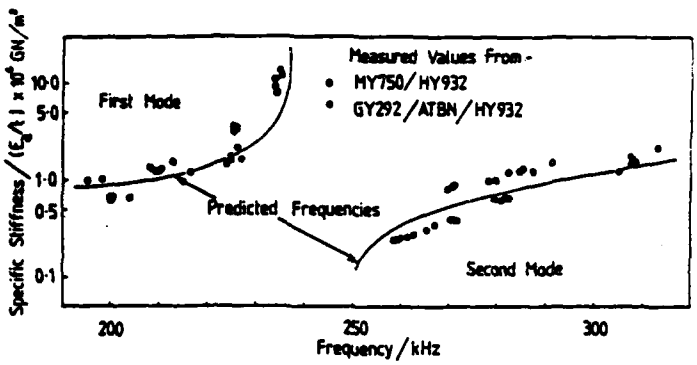


Fig 1 Predicted and measured resonant frequencies using the Fokker bond tester (From Guyott *et al*[8])

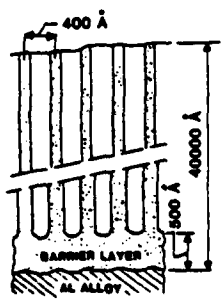
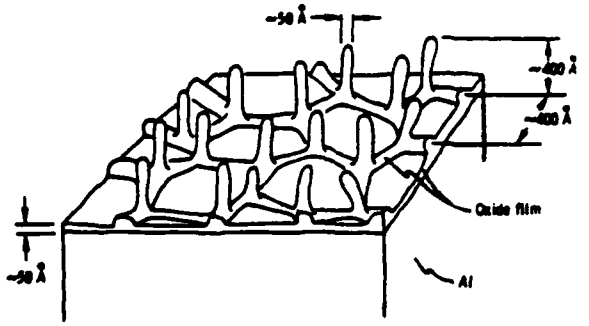
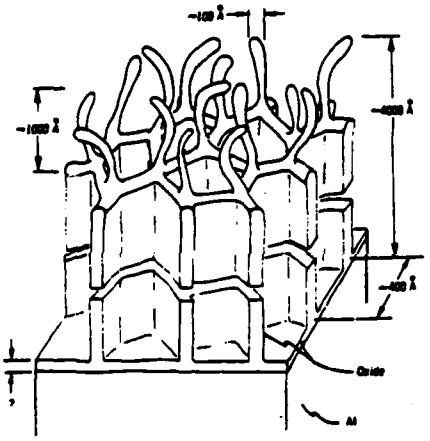


Fig 2 Schematic representation of the pore structure resulting from the European chromic acid anodising process (From Kwakernaak *et al*[13])

Figs 3-5



(a) The FPL Process – chromic acid etch



(b) A phospheric acid anodising process

Fig 3 Oxide morphologies produced on an aluminium surface by two different processes (From Venables[7])

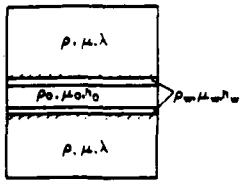
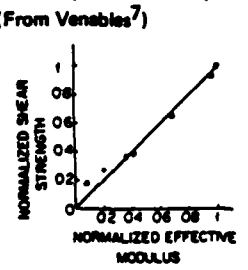


Fig 4 Rokhlin's multilayer model *of a bond line with weak* boundary layers



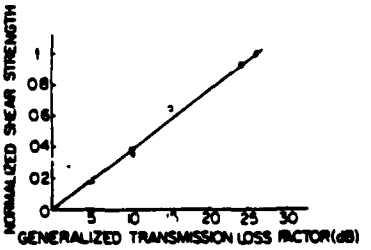
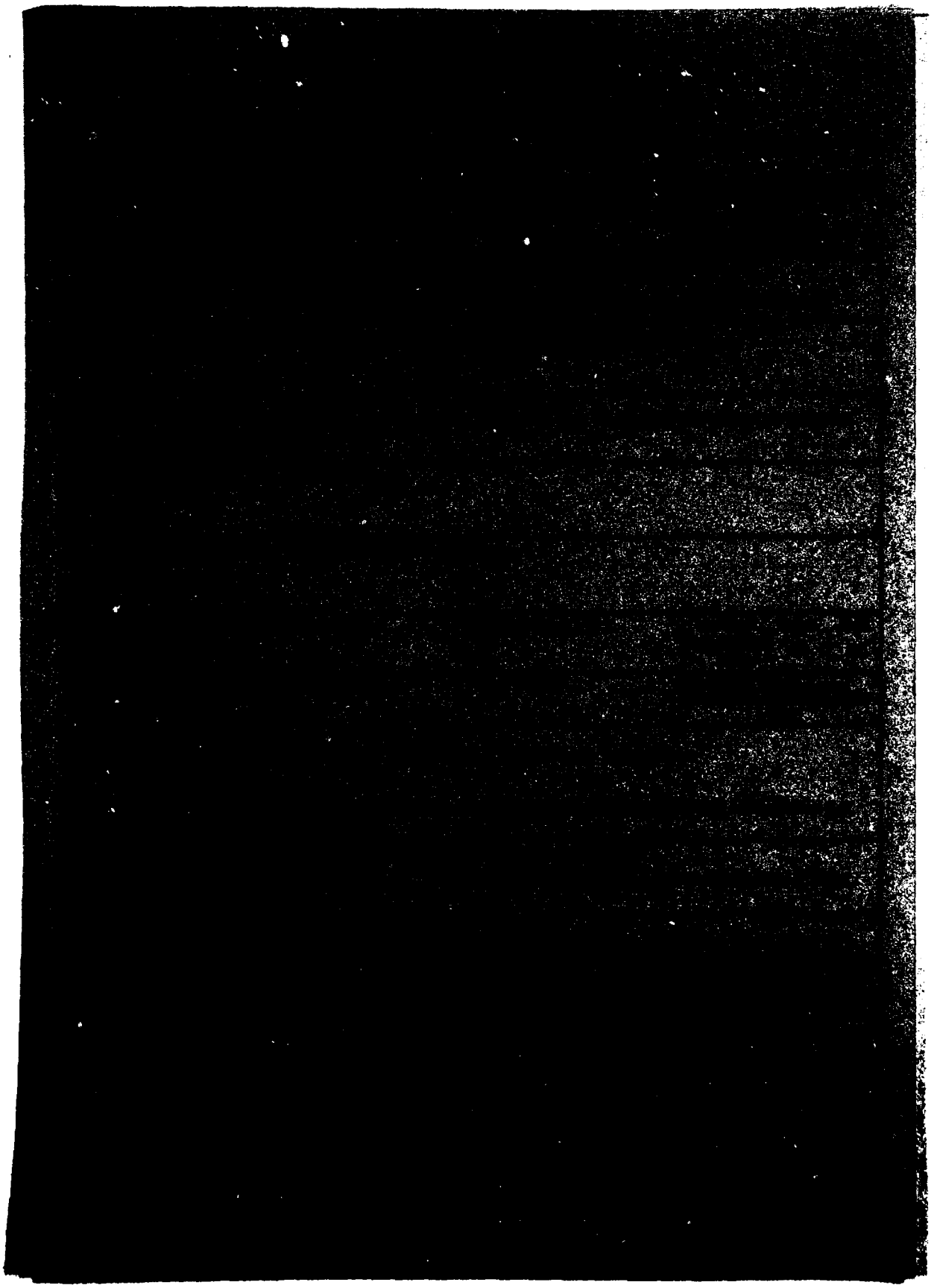


Fig 5 Relationship between the normalised shear strength and
(a) Effective shear modulus
(b) Generalised transmission loss factor
(From Rohklin *et al*[18])



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