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# The three-fibre method in determining environmental resistance of a fibre–resin bond

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The three-fibre method used for determining the adhesion between glass fibres and resin, has proved suitable for measuring the environmental effects on the bond strength. The environment used was hot water (95° C) treatment, the decreasing effect of which on the strength was not very clear. This method has two great advantages; it measures the actual bond strength and the environmental effect is directed efficiently on the bond. In this form, the method is only suitable for thermoset resins.

## 1. Introduction

The environmental resistance of polymer–matrix glass fibre composites has for the past few years been the subject of intensive research in several laboratories. Apart from short-term strength, environment and time should be taken into account in discussing the strength of polymer–matrix composites. In studying the environment, attention should be paid to the chemical composition and temperature of the environment.

As far as strength is concerned, most essential in the structure of a composite is the interface between fibre and resin. Theories of the fibre–resin interface vary broadly, but all of them suggest a layer of integrated primary bonds on the interface [1]. Theory and practice have shown that only primary bonds ensure sufficient composite strength in adverse environments [2, 3].

One objective of environmental resistance experiments is to find out long-term changes in properties by short-term experiments, via the hardening of environments. The greatest problem in these experiments is to find out the correlations between the intensified treatments and normal circumstances. Defining the effective interrelations between different factors is especially difficult.

### 1.1. Environmental resistance and test method

In practice, environmental resistance consists of the resistance of a composite's different components and that of their interfaces to external factors. The outer interfacial surface normally consists of pure matrix, which thus protects the fibres and especially the inner fibre–resin interfaces from the direct effects of ambient chemical atmosphere. In the case of polymer–matrix composites, we should take into account the plastic's capacity to absorb and permeate different substances in gaseous and liquid form, which is greatly dependent on the temperature, pressure and time.

The environment consists mainly of chemicals, temperature and different kinds of radiation. In normal outdoor atmosphere, the most problematic factors are water and different impurities in the air, the composition and amount of which are dependent upon geographical location. Low and high temperatures, temperature shocks and, especially in humid circumstances, passing the freezing point (0° C) are problematic. The most extreme experiments include boiling the composites, which under natural conditions is usually out of the question. In natural circumstances, the only kind of

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radiation which is energetic enough to break the structure of the plastic material is ultraviolet (UV) radiation. Other kinds of radiation in nature are too low in energy, or their amount is too small to have any serious effects whatsoever.

In principle, the effects of different factors can be studied separately or jointly. A typical example of the latter method is the weatherometer, where temperature, humidity and the amount of UV radiation can be altered or varied periodically. The purpose of weatherometer experiments is to create controlled circumstances for defining the effects of outdoor atmosphere on the properties of the composite.

Experiments can be accelerated by making the circumstances more extreme [4, 5]. As we noted above, the acceleration achieved by this method is difficult to evaluate. Typical means for creating more extreme conditions, e.g. in the weatherometer, are increasing the energy of the radiation that the material is exposed to and making the radiation continuous. The amount of energy received by the object can then be calculated and compared with the total energy the object would receive during a certain period in normal conditions. The effects of the daily cycle can be studied with accelerated temperature variation cycles. Comparing the results of radiation and temperature cycle experiments with normal circumstances is relatively easy, whereas increased temperature and humidity are much more problematic.

Quite often the aim of study is to find out the nature of the adhesion in a composite interface, or to compare different material and treatment combinations. In these cases, extreme methods of treatment which are known to have rapid and marked effects can be used. The most well-known among these methods is the boiling experiment, in which the remainder strength of the dried composite is measured as a function of boiling time. This method can be used to distinguish between adhesions caused by primary and secondary bonds.

Standardized test pieces are commonly used in environmental experiments, which can indeed be recommended. Results obtained are free of the previously mentioned problems, and comparing the results with other test results is easy.

## 2. Experimental details

This study was aimed at determining the applicability of the so-called single bond test method for defining environmental resistance. The method used was the three-fibre method, the principle of which is shown in Fig. 1, and which is described in more detail by Järvelä *et al.* [5]. The method consists of forming a controlled joint (a drop) between the vertical fibre and the horizontal fibres. The drop of matrix is hardened, after which it is tested either directly, or exposed to controlled external factors before testing. In the test, the vertical fibre is drawn off from between the fixed horizontal fibres. The fibre thicknesses have been determined in earlier studies; on the other hand, the fibre thickness does not seem to have any essential effect on the results.

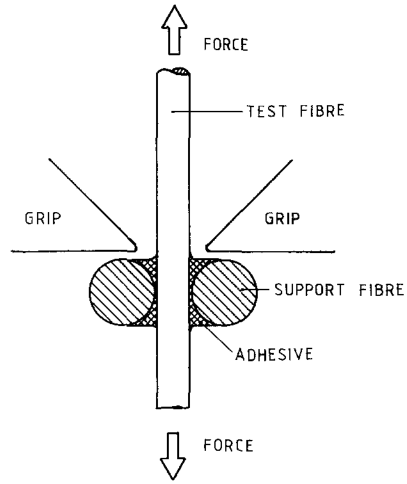


*Figure 1* A diagram of the three-fibre bond used for testing individual bonds.

This method can be expected to have the following advantages as compared to conventional methods. The three-fibre method has proved suitable for determining the actual binding strengths between glass fibre and different matrix materials. The bond used has such small dimensions that it is exposed to the full influence of the environment. The bond has an open fibre–binding material interface, through which the external factors can penetrate the actual interface. Due to its small dimensions, the interface follows the temperature of environment without great delay, and the formation of thermal gradients which would break the structure is prevented. Liquid and gaseous substances can rapidly diffuse through the binding material matrix into the interface, which decreases the occurrence of large concentration gradients in the bond.

All in all, this method can be considered a hardened environmental test on a small, separate bond; the hardening is achieved by the method, not by adverse environments. Furthermore, it should be noted that this method is relatively rapid, and does not, except for the tensile grips, require special laboratory equipment.

## 2.1. Test materials, conditions and results

The sole purpose of this work was to investigate the applicability of the three-fibre method for environmental tests. For this reason, materials were selected on the basis of work [5] to facilitate experimenting. Commerical glues were selected as materials: Loctite glass glue and Super Epoxy two-component epoxy glue. The fibre material used was A-glass fibre drawn in laboratory conditions and without surface treatment. Attempts were not made to remove the layer of water absorbed on the fibre surface in normal atmospheres.

The environmental treatment that the test materials were subjected to was a modified boiling test, where the bonds were kept submerged in water in a weatherometer at a temperature of 95° C. This experiment is here called a boiling test, although this may give an erroneous impression.

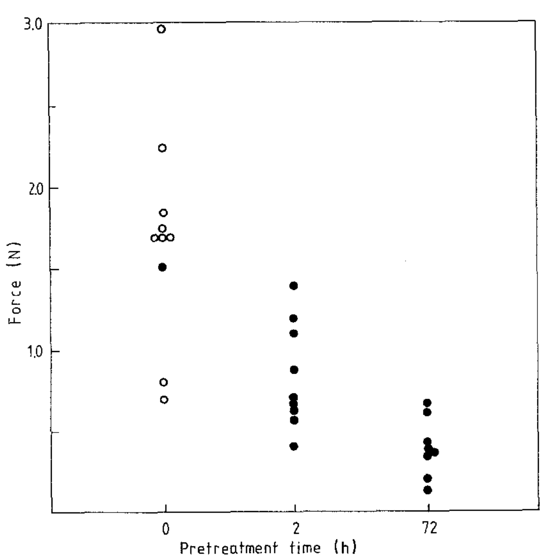


*Figure 2* The effects of the hot water treatment (water temperature 95° C) on breaking forces in the case of a two-component epoxy resin (Super Epoxy glue).

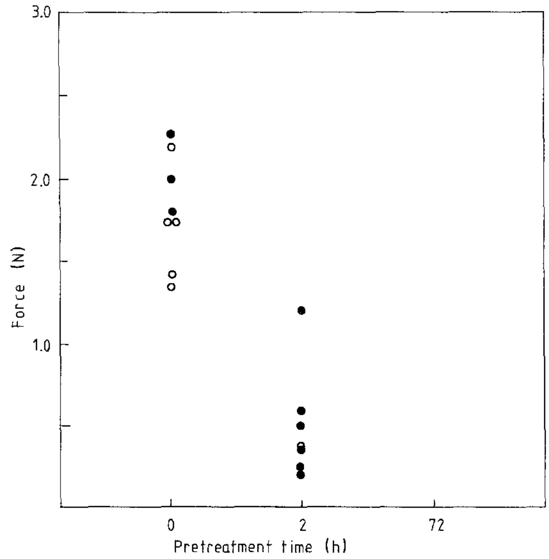


*Figure 3* The effects of hot water treatment (water temperature 95° C on breaking forces in the case of Loctite glass glue.

The treatment times were 2 and 72 h or only 2 h. The results were compared with the strengths of untreated bonds. A treatment period of 72 h is in any case so long that the deterioration of results is evident almost without exception. Figs. 2 and 3 show the force necessary for breaking the bonds as a function of time. Bond strength can be calculated on the basis of the breaking force according to the geometries presented in Fig. 1. Breaking force and bond strength correlate directly; they can therefore be used here to denote the same thing.

After the different treatments, the broken bonds were subjected to scanning electron microscopic (SEM) investigation. Fig. 4 shows a picture of both vertical fibres and of the hole that the detachment of the vertical fibre caused in the case of epoxy resin after the 2 h water treatment at 95° C.

## 3. Discussion

To start with, the results can be studied as a function of environmental treatments. Where Super Epoxy was used as binding material, we notice that, in most untreated bonds, the break occurred in the fibre. Thus the strength of the bond was greater than the results indicated. This is a disadvantage in the three-fibre method because, to succeed, it requires a strong vertical

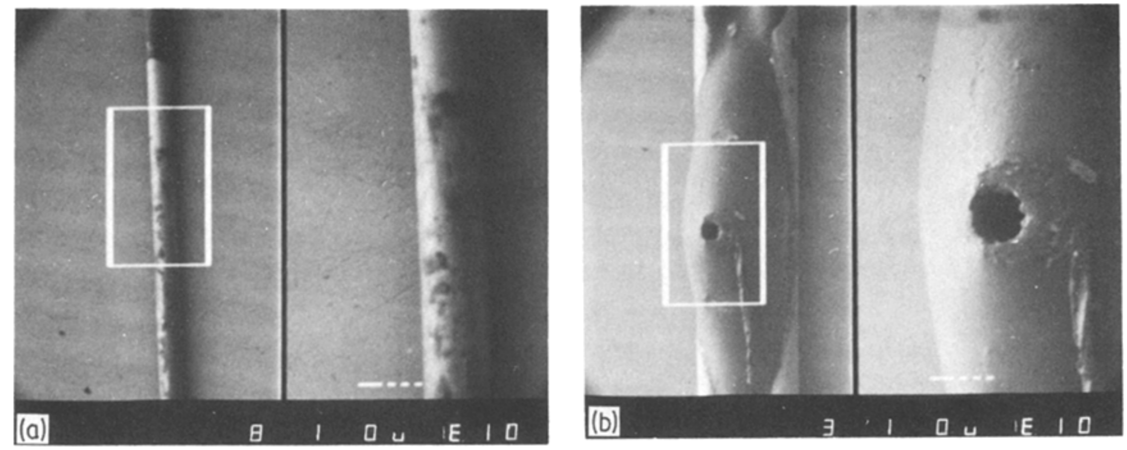


*Figure 4* SEM photographs of (a) vertical and (b) horizontal fibres after 2 h treatment in water of temperature 95° C (Loctite glass glue). Right-hand sides are magnified (× 3) views of area marked on the left-hand sides. *Long* white scale bars = 100 $\mu$m.

fibre. With bonds subjected to environmental treatment, it should be noted that all the breaks occurred in the bonds; thus the values obtained were clearly lower than those of untreated bonds. Table I summarizes, for both binding materials, the maximum, minimum, average, and average dispersion values of breaking forces indicating the strength of the bond. In the case of Super Epoxy, the environmental treatment decreased all these values. A very interesting finding was the clear decrease in the dispersion of results, which can be interpreted as a result of a break in the bond between the fibre and the matrix.

Where Loctite glue was used as the binding material, boiling caused a clear decrease in strength, as presented in Fig. 3. Fig. 4 illustrates with SEM photographs the fracture surfaces of the bond. The bond is represented with two photographs; the first shows the support fibres and the resin drop and the second shows the test

TABLE I The forces to break the bonds in the three-fibre method.

| Forces (N) | Loctite glass glue treatment time (h) | | Epoxy resin (Super Epoxy) treatment time (h) | | |
|---|---|---|---|---|---|
| | 0 | 2 | 0 | 2 | 72 |
| Maximum | 2.22 | 1.18 | 3.05 | 1.42 | 0.65 |
| Minimum | 1.40 | 0.23 | 1.70 | 0.39 | 0.14 |
| Average | 1.83 | 0.52 | 1.94 | 0.84 | 0.39 |
| Standard deviation | 0.31 | 0.35 | 0.49 | 0.34 | 0.18 |

fibre. The SEM photographs have been taken with a scanning electron microscope ISI 40 SEM provided with an anticontamination system, which allowed the examination of electrical nonconductive probes without coating.

Microscopic photography revealed that in the case of Loctite glass glue the binding material stuck partially to the fibre, whereas Super Epoxy showed no signs of this. In the case of untreated materials, the breaking surfaces of the binding material were more irregular.

## 4. Summary

The purpose of this work was to investigate the applicability of a certain microscale method in determining the environmental resistance of a bond. The method has previously proved suitable for determining the actual strength of a bond. The method used was the so-called three-fibres method.

Environmental treatments were water treatments carried out at a temperature slightly below boiling temperature. The effects of the environmental treatment were clearly visible; the main part of the results can be explained by the combined effect of treatment and treatment time. These results also lend strong support to using this method to determine bond strengths, because the treatment can be expected to decrease the interface strength of a bond, in particular. This is clearly reflected in the results, which implies that the method also measures the bond strength.

A great advantage of this method is that the influence of the environment is directed efficiently on the actual bond. Different delays and concentration gradients are in this case minimized. The method can efficiently keep trace of the influences of different factors.

This method to measure environmental resistance can be regarded as an accelerated environmental test, where the acceleration is achieved by developing the method and not by extreme conditions. We can conclude that this method has proved useful in determining bond strength and its environmental resistance as a function of time.

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