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# Non destructive evaluation of adhesively bonded composite structures using high frequency dielectric spectroscopy

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Over the last ten years, the application of high frequency dielectric techniques for the assessment of adhesively bonded structures has been investigated. The technique has been used for the study of adhesively bonded aluminium structures and its application to carbon fibre reinforced plastic (CFRP) bonded structures forms the basis of this paper. The electrical conductivity of the carbon fibres in the CFRP composite materials is sufficiently high for adhesively bonded structures to exhibit the properties of a wave-guide. The non-conductive adhesive behaves as a dielectric. The time domain data allows the integrity of the structure to be explored and is sensitive to the orientation of the fibres at the adherent-adhesive interface. Furthermore, a good correlation is shown between time domain dielectric spectroscopy and gravimetric results. This study indicates that the success obtained in the application of high frequency dielectric measurements to adhesively bonded aluminium structures is also applicable to CFRP bonded structures. The dielectric studies not only indicate a new way to assess the state of such a structure but also are producing new insight into the application of dielectric time domain response (TDR) measurement to non-isotropic materials. 

## 1. Introduction

The integrity of power and signal cables and related structures is typically assessed by the use of time domain electrical measurement techniques. In the past fifteen years development of vector network analysers operating in the frequency range 300 kHz to 3 GHz has allowed accurate measurements of the electrical permittivity and loss in wave-guide structures.

During the past decade, the application of high frequency dielectric spectroscopy to the assessment of aluminium adhesively bonded structures has been investigated. It has been established that information on the quality and integrity of bonded structures can be obtained [1–5]. Its potential as a non-destructive technique to monitor ageing and degradation of adhesive joint structures during exposure to a harsh hot and humid atmosphere has been demonstrated [3]. Time domain analysis allowed identification of defects present in the bond line [2–4], whereas frequency domain analysis allows the effect of the ingress of water in the bond line [5] to be assessed.

Degradation of bonded structures occurs when the adhesive absorbs significant amounts of moisture. Once the water penetrates the bond line, the adhesive properties can be altered in a reversible manner by plasticisation or in an irreversible manner by hydrolysis or crack and craze formation [6, 7]. Generally, degradation at the adhesive-adherent interface occurs by displacing the adhesive or by changing the adherent surface chemistry. Furthermore, the diffusion of water in the adhesive tends to modify the mechanical failure mechanism of the joint from a cohesive failure in the adhesive layer to an adhesive failure at the adhesive-adherent interface [8, 9].

High frequency dielectric spectroscopy techniques have proved successful in the study of ageing of aluminium-epoxy resin-aluminium bonded structures since the conducting aluminium adherent generates a

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wave-guide structure for the propagation of electromagnetic waves. However, the aerospace industry increasingly uses adhesively bonded composite materials in aircraft primary and secondary structures. There is, therefore, a requirement for the development of non-destructive techniques to assess the integrity of these bonded composite structures. This paper explores the potential of high frequency dielectric spectroscopy as a non-destructive evaluation (NDE) method for adhesively bonded composite structures. This paper also addresses how electromagnetic wave propagation may occur in composite structures.

## 2. Experimental procedure

### 2.1. Materials

The carbon fibre reinforced plastic (CFRP) adherents were manufactured from Hexcel Composites Ltd unidirectional carbon fibre pre-impregnated film, trade named 914C-TS(6K)-5-34%. The pre-impregnated film contained 66% in weight of high-tensile surface treated carbon fibres. The adhesive system used for bonding the CFRP plates was a 3M structural epoxy system trade named Scotch-Weld Brand AF-163-2U, consisting of a nylon woven fibres supported epoxy resin.

### 2.2. Manufacturing process

Carbon fibre pre-preg layers were used to produce a series of different lay-up designs using a vacuum bag procedure in an autoclave. Curing was achieved at a temperature of 170°C under a pressure of 7 bar. A post-cure at 190°C for 4 hours was performed. Joints were manufactured by joining two carbon fibre reinforced plastic (CFRP) plates with a lay-up of adhesive films. The assemblage, placed in an autoclave chamber, was cured using a vacuum bag system at a temperature of 125°C and a pressure of 2 bar for 1 hour. The final system was a single lap joint made of 150 mm length, 50 mm width and 1.8 mm thick CFRP plates bonded over a 10 mm wide overlap by a 1 mm thick adhesive.

### 2.3. Gravimetric measurements

As soon as manufactured, all the samples were placed prior to their ageing in a dessicator at room temperature in order to avoid absorption of moisture. Gravimetric measurements during ageing were performed by removing the samples from a water bath held at a constant temperature of 60°C and rapidly blotted and weighed using an electronic balance (Mettler AJ100) with an accuracy of ±0.1 mg. The times for the weighing experiments were assumed to be sufficiently short not to influence the values of the mass measured.

### 2.4. Dielectric measurements

Dielectric measurements were carried out in reflection mode over the frequency range 300 kHz to 3 GHz using a Hewlett Packard 8753A Network analyser. The system was calibrated using three independent standards whose reflection coefficients are known over the frequency range of interest: short, open circuit and a matched load at 50 Ω. Details of the measurement technique and theory have been presented elsewhere [1, 4]. The ratio of reflected to incident waves $\rho$ for such wave-guide line is given by the following equation

$$\rho(t) = \rho_i(t)\delta(t) - \left[(1-\rho_i)^2\rho_T \sum_0^N (\rho_i\rho_T)^{N-1}\delta(t-NT)\right] \quad (1)$$

where $\rho_i$ and $\rho_T$ are the reflection coefficients at the input and end of the joint respectively, $t$ is the time, $T$ is the total transit time, $N$ is the number of transits in a time $t$ and $\delta$ is the Kronecker delta. The reflection coefficients $\rho_i$ and $\rho_T$ are defined by

$$\rho_{\mathrm{i}} = \frac{Z_l - Z_0}{Z_l + Z_0} \qquad \text{and} \qquad \rho_T = \frac{Z_T - Z_l}{Z_T + Z_l}$$

where $Z_0$, $Z_l$ and $Z_T$ are the characteristic impedance of the system, joint and line termination respectively. The latter is equal to infinity for free end, therefore $\rho_T$ equals one. Time domain data were obtained by using the network analyser's inverse Fourier transformation over the frequency range.

## 3. Results and discussion

### 3.1. Joint design

When good contacts exist between the fibres of CFRP laminates, the composite adherent's structure is able to transmit electric current polarising the adherent/adhesive interfaces. In a previous study, joints were designed so that the unidirectional fibres were in the direction of the wave propagation (Fig. 1, top). The limitation of this design was that the water diffused perpendicularly to the mechanical testing direction, resulting in a limited understanding of the effects of water on the degradation of the mechanical properties of the bonded structure.

The aim of this study was to produce a design, which eliminated this restriction. In the first design investigated, the unidirectional fibres were orientated in the

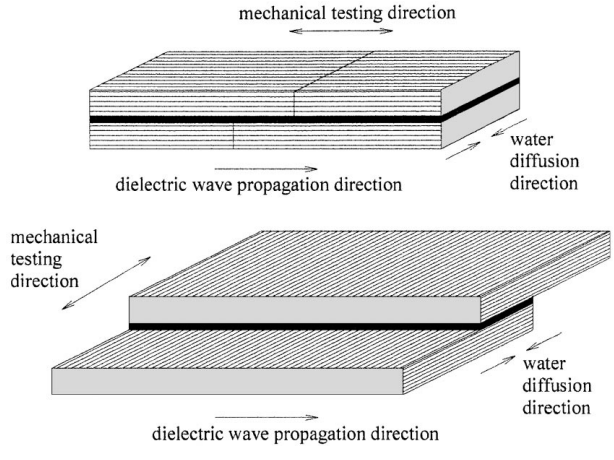



*Figure 1* Previous joint design (top) and new joint design concept (bottom).

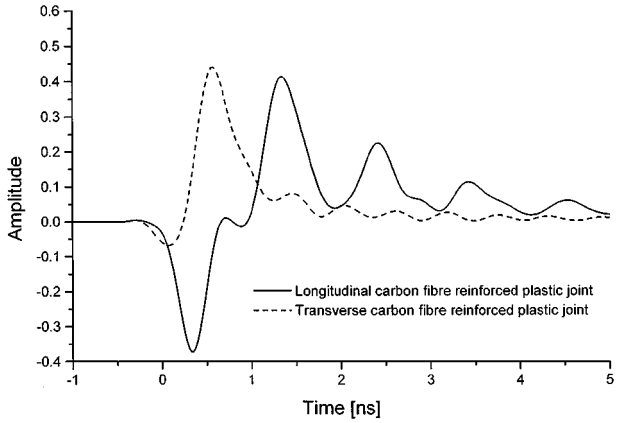



*Figure 2* TDR of longitudinal and transverse CFRP joints.

same direction as the water diffusion and mechanical testing directions; therefore perpendicular to the direction of the electromagnetic wave transmission (Fig. 1, bottom). Results for unidirectional carbon fibre plates joint are presented in Fig. 2 and several reflection peaks were observed (solid line). This is a typical TDR spectrum. When all the fibres are perpendicular to the dielectric wave propagation direction, the dissipation of the impulse energy in the first millimetres of the joint prevents the development of secondary reflection peaks. There is enough energy to percolate through the thickness of the plate, as shown by the presence of a first upward peak, but the high resistivity of the laminate matrix prevents propagation of the pulse along the joint.

The main issue following these preliminary studies was to improve the conductivity of the adherent in order to propagate the signal along the joint without significantly affecting the mechanical testing and involved orientation of the fibres in the propagation direction. During high frequency dielectric spectroscopy, an electric field **E** appears between the CFRP plates producing a magnetic field **H**, which propagate an electrical wave at the adherent/adhesive interface, see Fig. 3. This part of the theory has not been previously explored since only isotropic materials such as aluminium have been studied. By using composite materials, such as carbon fibre pre-pregs, there is the possibility to modify the adherent/adhesive interface by changing the orientation of the fibre of the laminae at the interface.

For this purpose, five different lay-ups following the sequences $[0_{14}]$, $[0_2/90/0_2/90/0]_S$, $[0/90_3/0/90_2]_S$, $[90_2/0/90_2/0/90]_S$ and $[90/0_3/90/0_2]_S$ were manufactured. The differences between these lay-ups, which modify the overall dielectric property of the adherent,

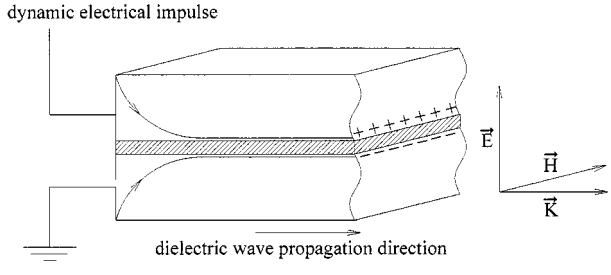
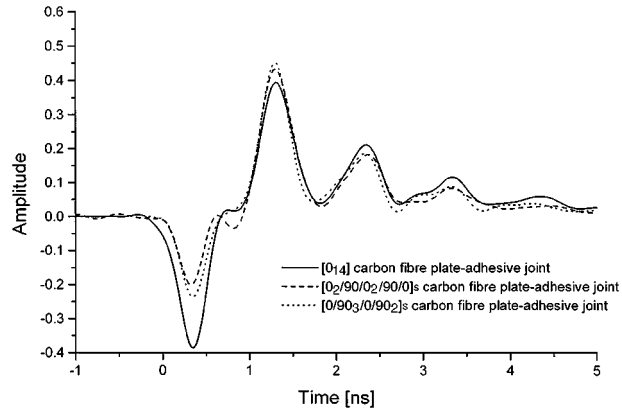
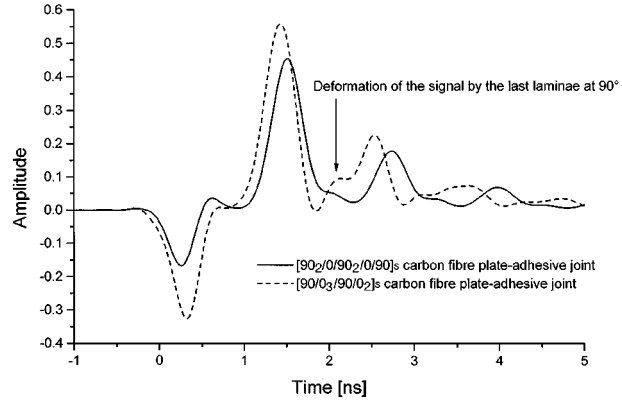



*Figure 3* Schematic representation of transverse electro-magnetic (TEM) wave propagation.




(a)




(b)

*Figure 4* TDR of different carbon fibre lay-up: (a) last laminae at the interface at 0° and (b) last laminae at the interface at 90°.

are designated in the direction of the last laminae at the interface and the percentage of fibres in each direction. The percentage of fibres in the 0° direction in each of the lay-ups is 100%, 74%, 26%, 26% and 74%, respectively. It should be noted that the directions of the carbon fibres are given relative to the dielectric wave propagation direction. A 90° fibre direction means that the fibres are perpendicular to the dielectric wave propagation (Fig. 1, bottom).

Fig. 4a presents the TDR spectrum of the $[0_{14}]$, $[0_2/90/0_2/90/0]_S$ and $[0/90_3/0/90_2]_S$ lay-up carbon fibre plate-adhesive joint (respectively 100%, 74% and 26% of the carbon fibre at 0°) with the direction of the fibre of the laminae at the interface being at 0°. Fig. 4b presents the TDR spectrum of the $[90_2/0/90_2/0/90]_S$ and $[90/0_3/90/0_2]_S$ lay-up carbon fibre plate-adhesive joint (respectively 74% and 26% of the carbon fibre at 0°) with the direction of the fibre of the laminae at the interface being 90°.

The sequence of the layers does not have an influence on the response and, as seen in Fig. 4b, the presence of some layers at 90° does not significantly disturb the response when comparing the $[0_{14}]$ response with the other laminate responses. For mechanical reasons, designs with fibres at 0° at the interface are not allowed since they will be at 90° for the shear test. Thus, from these results, the $[90_2/0/90_2/0/90]_S$ configuration was selected for the adherent plates of the new joint design, since it is the best compromise between all the experiment requirements. This implies 14 layers of carbon fibres, 10 at 90° and 4 at 0°, representing 74% of fibres

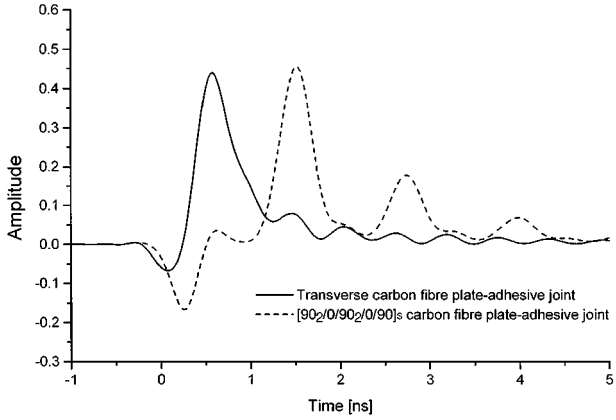
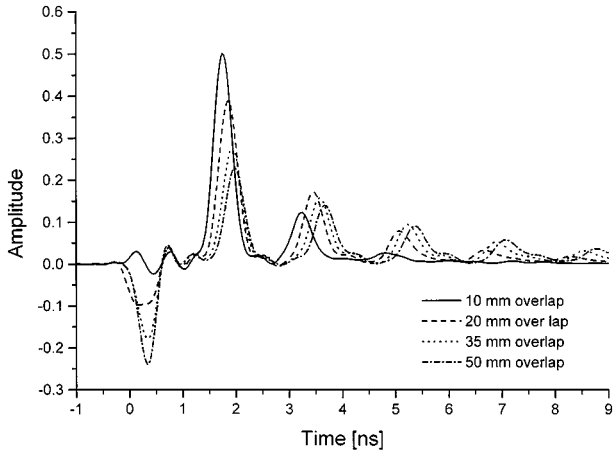


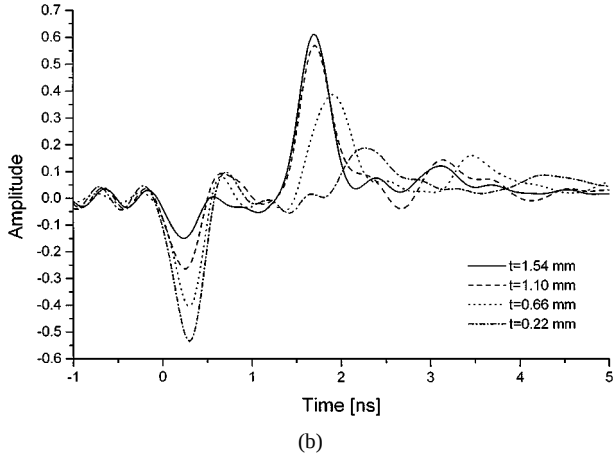

*Figure 5* TDR of $[0_{14}]$ and $[90_2/0/90_2/0/90]_S$ CFRP joints.

at 90° and 26% at 0°. Fig. 5 shows the response of the new joint design, where by inserting carbon fibre laminae in the longitudinal direction, the electric pulse is efficiently transmitted.

Dielectric spectroscopy of adhesive joints is mainly governed by two parameters: the electrical behaviour of the materials used in the joint and the joint geometry. As the former is constant for a given material, the only way to optimise TDR spectra is to modify the geometry of the joint. Practical requirements determined the length, width and thickness of the carbon fibre plates. Therefore, only the thickness of the adhesive and the width of the overlap of the joint were left to improve its dielectric response. Fig. 6a and 6b, showing the influence of the overlap dimension and adhesive thickness, respectively, on the TDR spectrum, demonstrate that the thickness and the overlap are in close relationship.




(a)




(b)

*Figure 6* Effect of (a) the overlap dimension and (b) the thickness of the adhesive on the TDR.

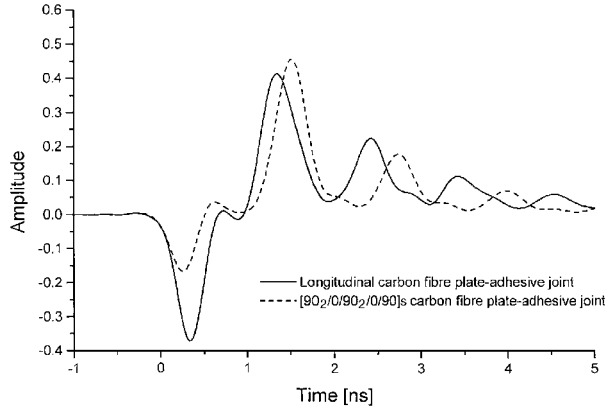



*Figure 7* Comparison of the TDR of the previous (—) and new designs (- -).

Indeed, in an infinite capacitor placed in vacuum, the characteristic impedance $Z_0$ is given by

$$Z_0 = \frac{t}{w} \cdot \sqrt{\frac{\mu_0}{\varepsilon_0}} \qquad (2)$$

where $\mu_0 = 4\pi \times 10^{-7}$ Tesla meter/Ampere is the vacuum permeability constant, $\varepsilon_0 = 8.854188 \times 10^{-12}$ Farad/meter is the vacuum permittivity, $w$ is the overlap dimension of the capacitor and $t$ the thickness of the insulator. In this study, the dielectric spectroscopy is performed with $Z = 50\,\Omega$. Consequently, the overlap dimension of infinite conductive plates over an infinite insulator must be 7.5 times the thickness of the insulator. For the CFRP-epoxy structure, this factor has been estimated at 10.

Therefore, the final dimensions chosen for the joint were 10 mm of overlap for 1 mm of adhesive thickness. As a result, the new joint design was made of carbon fibre plates 150 mm long, 50 mm wide and 1.8 mm thick with an adhesive of 150 mm length, 10 mm width (overlap) and 1 mm thickness.

Fig. 7 presents a comparison between the TDR of the previous design and new design. The lower amplitude in the impulse (downward) peak indicates a better balance between the impedance of the measuring system and the joint structure. The higher amplitude of the first response (upward) peak corresponds to an improved polarisation of the CFRP-adhesive interfaces. It is believed that the percolation between the fibres at 90° minimises the energy required for the electrical wave to reach the adherent-adhesive interfaces, consequently optimising the polarisation of the adhesive. The displacement of the peaks towards longer response time is consistent with the high resistivity of the CFRP matrix, which decreases the velocity of the Transverse Electro-Magnetic (TEM) wave.

## 3.2. Water uptake and high frequency dielectric measurement

### *3.2.1. Gravimetric measurements*

Fig. 8 presents the results for the joint structure, the CFRP adherents alone and the adhesive. The amount of water present in the adhesive has been estimated by

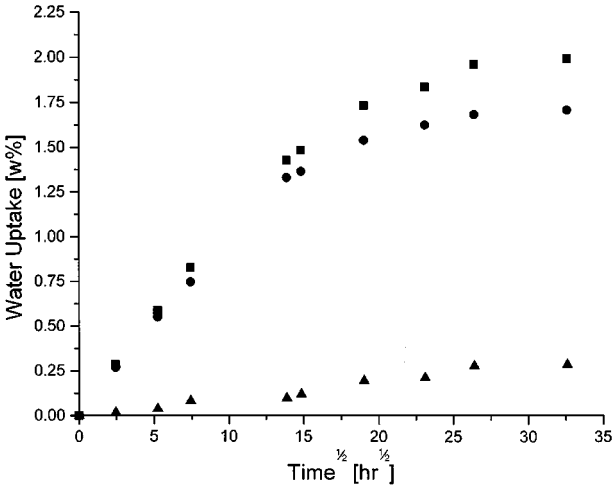



*Figure 8* Water uptake of (■) the joint structure, (●) the CFRP adherents and (▲) the adhesive.

subtracting the amount of water present in the joint to the amount of water present in the CFRP plates. The latter has been calculated from gravimetric results of CFRP plates exposed to identical ageing conditions. From these results, it is possible to dissociate the amount of water present in the adherent from the amount of water absorbed by the adhesive.

### 3.2.2. Effect of water on the TDR of CFRP plates

CFRP plates were manufactured according to the new design. Dielectric measurements were performed by creating an air gap similar to the adhesive geometry between the adherents using PTFE spacers at both ends of the plates. Several TDR measurements were carried out prior to ageing and a statistical analysis was applied to the results. Immersion of CFRP plates in tap water at 60°C for up to 1870 hours was followed by TDR measurements.

Fig. 9 presents a comparison of the TDR results before and after immersion. No significant difference can be observed between both measurements despite a water uptake of almost 2%. This indicates that any variations that may be observed in the TDR of adhesively bonded structures are caused by changes in the dielectric behaviour of the material present between the two wave-guides, in this case the CFRP plates.

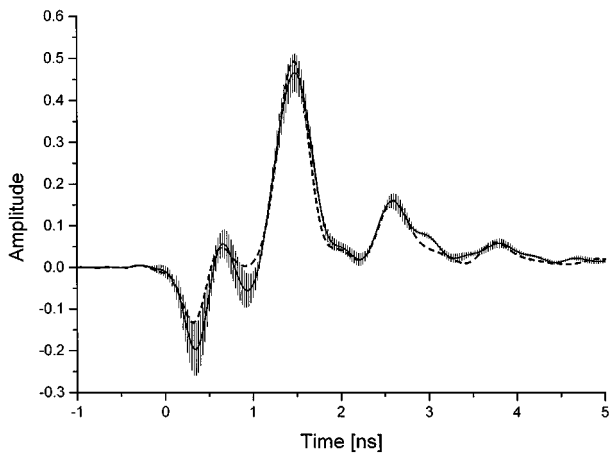



*Figure 9* TDR of CFRP plates: unaged (—) and after 1870 hours at 60°C in tap water (- - -).

Therefore, the penetration of water in the waveguides, which may alter their dielectric properties, does not affect the TDR results. Since alteration of the characteristics of polymeric materials by plasticisation, hydrolysis or crack and craze formation significantly change their dielectric properties, TDR analysis of high frequency dielectric measurement should provide an efficient non destructive method to assess the degradation of adhesively bonded composite structure. For similar reasons, delamination at the adherent-adhesive interface could be observed by TDR measurements.

### 3.2.3. Effect of water on the TDR of adhesively bonded CFRP plates

In a polar gas, where the density of molecules is low, the static dielectric permittivity $\varepsilon_s$ is related to the dipole moment $\mu$ of a molecule by Debye's equation [10]

$$\frac{\varepsilon_s - 1}{\varepsilon_s - 2} = \frac{4}{3}\pi N\left(\alpha + \frac{\mu^2}{3kT}\right) \tag{3}$$

where $k$ is the Boltzman constant, $T$ is the temperature, $N$ is the number of molecules per unit volume and $\alpha$ is the 'deformation' polarisibility which arises due to the elastic displacement of the molecules. This expression is however inadequate for description of the response of condensed materials because it neglects both intramolecular and intermolecular bonding and assumes the local electrostatic field acting on the dipoles is zero. The Kirkwood-Fröhlich equation allows for the effects of the local field and has the form [11, 12]

$$\frac{(\varepsilon_s - \varepsilon_\infty)(2\varepsilon_s + \varepsilon_\infty)}{\varepsilon_s(\varepsilon_\infty + 2)^2} = \frac{4\pi N}{9kT} g\mu_0^2 \tag{4}$$

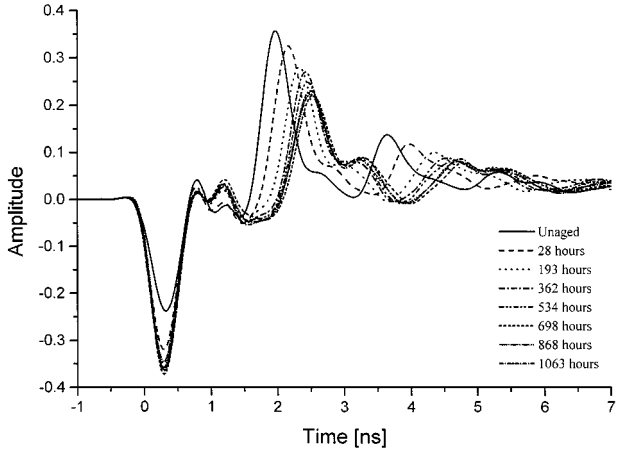
where $\varepsilon_\infty$ is the dielectric permittivity at a frequency so high that the dipole orientation contributions have vanished and $g$ is the orientation correlation function, which is a measure of the local ordering in the material and is defined as

$$g = 1 + \sum_{\substack{j=1 \\ j \neq i}}^{N_0} \overline{\cos \gamma_{ij}} \tag{5}$$

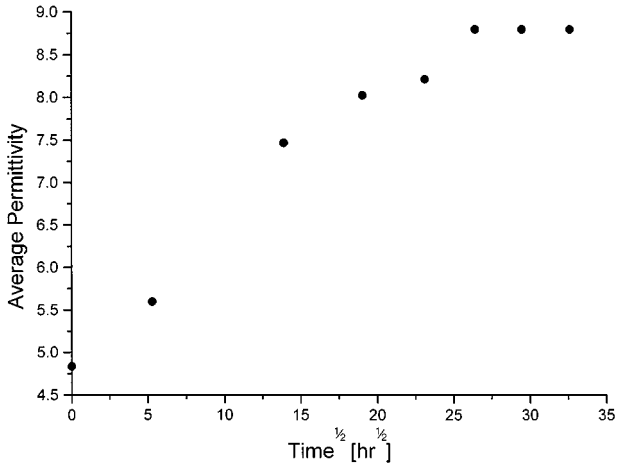
with $\overline{\cos \gamma_{ij}}$ being the average of the cosine of the angle $\gamma$ made between the dipole $i$ and $j$, and $N_0$ is the number of elementary dipole units contained in the spherical region considered.

Therefore water uptake could be assessed by assuming that the variation in the dielectric permittivity of a material during exposure to water is caused by a variation in the number of dipole present, mainly water dipoles [13]. Previous work [2, 14] has shown that water penetration in polymeric materials can be measured by dielectric spectroscopy.

TDR measurements were performed during ageing and are presented in Fig. 10. The time difference between two neighbouring peaks is related to the




(a)




(b)

*Figure 10* Evolution of (a) the TDR and (b) the average permittivity during ageing.

effective permittivity $\bar{\varepsilon}$ of the material between the wave-guides by [3]

$$\bar{\varepsilon} = \left(\frac{c}{2 \cdot l/\Delta t}\right)^2 \tag{6}$$

where $c$ is the velocity of light, $l$ is the physical length of the joint, $\Delta t$ is the time difference between the impulse peak and the first response peak and $\bar{\varepsilon}$ is the average dielectric permittivity.

Fig. 10a shows the displacement of the response peaks towards longer response time values. The change of the dielectric permittivity of the adhesive due to the presence of water decreases the electrical wave velocity, therefore displacing the response peaks towards longer time response values. Fig. 10b presents the variation of the average dielectric permittivity as a function of exposure time. The TDR measurement also provides information on the quality of the bond line. It is believed

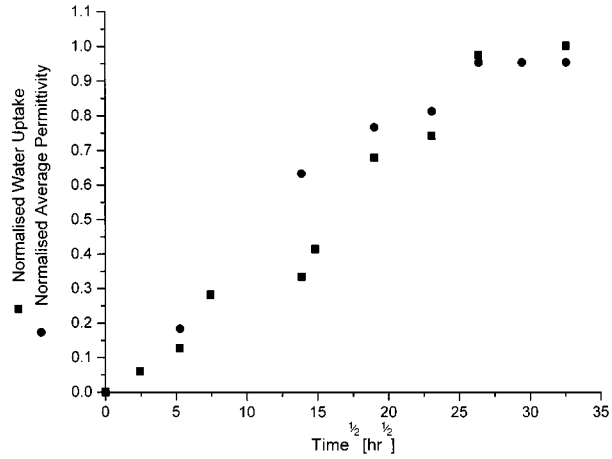



*Figure 12* Correlation between normalised adhesive water uptake and average dielectric permittivity.

that the second upward small peak, at 1.225 ns, is due to macro-voids present in the adhesive bond line as can be seen in Fig. 11 which is an ultra-sonic C-Scan of the joint where the lighter areas in the grey zone represent voids in the bond line.

When the electrical wave reached these defects, the change in the dielectric permittivity generated a reflected wave, which produced a weak response peak. The rapid increase of this peak amplitude to an equilibrium value is due to the filling by water of the voids.

### 3.2.4. Correlation between water uptake and TDR of adhesively bonded composite structures

In order to compare the dielectric frequency domain and time domain results with the gravimetric results, normalisation of the data was performed using Equation 7

$$\text{Data}_{\text{Norm}} = \frac{\text{Data}_t - \text{Data}_0}{\text{Data}_\infty - \text{Data}_0} \tag{7}$$

where $\text{Data}_t$ is the value at time $t$, $\text{Data}_0$ is the initial value, $\text{Data}_\infty$ is the final value at equilibrium and $\text{Data}_{\text{Norm}}$ is the normalised value.

A correlation coefficient of 0.96 between the TDR results with the gravimetric results shown in Fig. 12 confirms that water penetration in adhesively bonded structure could be assessed by high frequency TDR measurement.

## 4. Conclusion

This study has shown that water ingress in non-isotropic adherent used as wave-guides has no effects on time domain high frequency dielectric analysis. Correlation

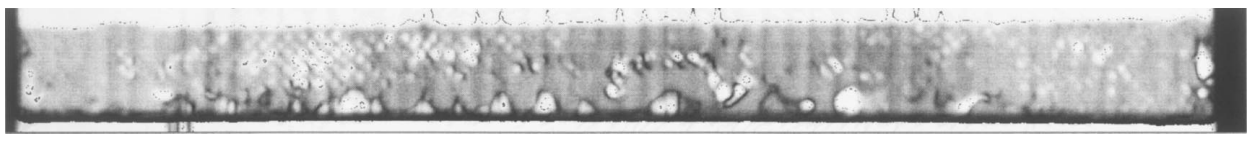


*Figure 11* Ultra-sonic C-Scan of the adhesively bonded joint.

between the TDR and gravimetric data validated the use of TDR to identify the change in the dielectric permittivity of the bond line and to relate it to the water uptake in the structural adhesive. Furthermore, the analysis of the TDR spectrum can also provide information on the quality of the bond line and the presence of defects.

This study on adhesively bonded composite structure has shown the potential of high frequency dielectric TDR as a NDE method to assess the integrity of the bond line present in aircraft primary and secondary structures.

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