

A review of NDE methods for porosity measurement in fibre-reinforced polymer composites

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Low porosity levels are essential for ensuring the performance of carbon fibre-reinforced composite structures. Despite many attempts to measure absolute porosity levels as a process-control tool, the currently accepted methods often rely on relative measurements in comparison with a reference standard. However, newer low-cost manufacturing methods such as resin-transfer moulding (RTM) and resin film infusion (RFI) would benefit from a reliable absolute 'porosity meter.' This paper reviews the literature documenting NDE methods for porosity measurement in fibre-reinforced polymer composites. The majority of the papers have concentrated on ultrasonic methods to determine porosity levels, mainly for quality control purposes. A few workers have reported on the application of other NDE methods. The review concentrates on methods that have potential for use during composite manufacture. To date, no one NDE method has been able to provide a porosity measurement independent of both the pore morphology and the fibre and resin matrix materials.

Introduction

There are several types of defect that can occur during the manufacture of carbon fibre-reinforced composites. One of the most serious is voids in the matrix, which can be further classified as:

- Delaminations: these are large planar voids occurring at the interfaces between the plies. They are easily detected by ultrasonic methods as they act as almost perfect reflectors of the ultrasonic beam.
- Discrete voids: these are large enough to be of structural significance and can also be individually detected and measured by ultrasound.
- Porosity: this can be described as a large number of microvoids, each of which is too small to be of structural significance or to be detected individually by a realistic inspection technique, but which collectively may reduce the mechanical properties of the components to an unacceptable degree. It is usually produced during the curing cycle from entrapped air, moisture or volatile products.

The distinction between discrete voids and porosity is a matter of convenience but for practical purposes, porosity may be thought of as sub-millimetre voids whereas voids of several millimetres dimension would be considered as discrete defects.

The occurrence of high levels of porosity has been recognised as a serious problem for composite materials for many years. As the compressive and interlaminar shear strength of carbon fibre-reinforced composites depend primarily on the matrix properties, these are reduced by the presence of porosity. In contrast, tensile properties, which are determined almost exclusively by the fibre

properties, are relatively unaffected by the presence of porosity^[1]. An extensive review of the effects of porosity on the mechanical properties of composites was undertaken by Judd and Wright^[2] and subsequent work has been reported by Uhl *et al*^[3]. These studies have concluded that the interlaminar shear strength is more seriously affected by the presence of porosity than the compression strength. It has generally been found that the interlaminar shear strength decreases by about 7 % per 1 % of voids, up to a total void content of about 4 %. Other mechanical properties are also affected, although not to the same degree.

A figure of 2 % porosity has commonly become the nominal acceptance threshold for many composite components. As an example, Hagemmaier and Fassbender^[4] state that for secondary aircraft structure, composite with void contents in excess of 2 % are unacceptable. Research programmes in NDE have therefore concentrated on providing techniques that could detect and measure porosity in the range of 1 to 5 % by volume. Ideally, an NDE technique is required that could determine the level of porosity in a composite laminate independent of other variables such as pore morphology and the fibre and matrix materials. However, to date, no one NDE method has been able to provide this universal 'porosity meter.' The majority of the work on porosity measurement has concentrated on ultrasonic techniques. This paper reviews these in some detail but also describes some other methods that have been proposed for porosity measurement. It should be noted that this review has concentrated on methods that have potential for the development of in-service techniques.

Review of ultrasonic methods for porosity determination

Ultrasonic velocity and attenuation can both be used to estimate the porosity level in composite material. The velocity and attenuation of an ultrasonic pulse travelling in a fibre-reinforced composite will be dependent on both the porosity (void fraction) and the fibre volume fraction of the composite. Measurements of attenuation have been used in many studies as it is simpler to measure and less affected by variation in the volume fraction of fibres in the reinforcement.

Both velocity and attenuation measurements depend on the frequency of the ultrasound used for the inspection. Williams *et al*^[5] studied the variation of attenuation and velocity with frequency in carbon composite. Longitudinal velocity was found to be only very weakly frequency dependent while the attenuation increased significantly with increasing frequency. In principle, the visco-elastic attenuation in matrix resin should increase linearly with frequency for low frequencies. This should be true for composite material at practical inspection frequencies (0.5 to 10 MHz), where the ultrasonic wavelength (6 to 0.3 mm) is much greater than the diameter of the reinforcing fibres.

Stone and Clarke^[6] carried out one of the earliest investigations into ultrasonic methods for porosity measurement at the Royal Aircraft Establishment (RAE), Farnborough. They carried out an extensive series of experiments on a set of panels made from prepreg material with HTS carbon fibre and ERLA 4617/DDM resin

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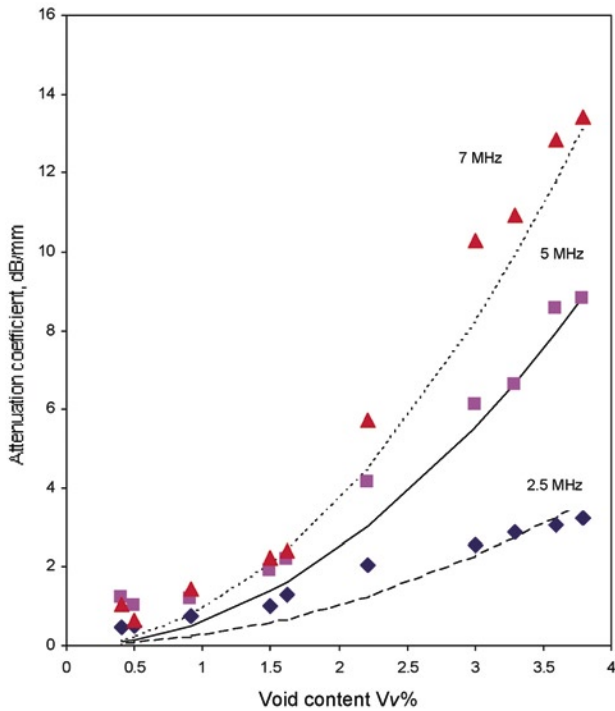


Figure 1. Variation of attenuation coefficient with void content^[6]. The curves show the fit to the data using equation 1

matrix, giving nominally 60 % fibre volume fraction. The panels were manufactured in an autoclave to a nominal thickness of 2 mm using a unidirectional lay-up. An attempt was made to vary the porosity in the panels by altering the pressures used during the cure cycle. Porosity values were obtained by destructive examination using acid digestion. The panels were found to have porosity in the range of approximately 0.4 to 4.0 %, with an estimated measurement error of ± 0.5 %. Both surfaces of the panels were ground flat so that all the panels were of constant thickness and surface finish. All the ultrasonic measurements were taken with the ultrasound propagating perpendicular to the carbon fibres.

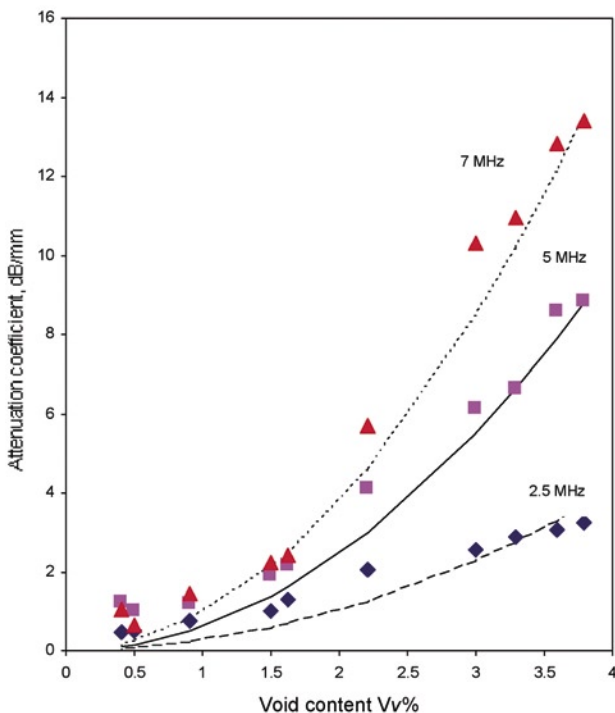


Figure 2. Variation of attenuation coefficient with void content^[6]. The curves show the fit to the data using equation 2

Stone and Clarke investigated the use of ultrasonic velocity measurements for determining void content. The velocity of this material was measured by the authors to be 2980 ms^{-1} . They calculated the time delay between the echoes from the front and back faces of a 2 mm-thick laminate to be $1.342 \mu\text{s}$. Experimentally they found that a void content of 5 % reduced the velocity by about 6 %. Therefore, a specimen with 1 % voids would have a velocity of about 2944 ms^{-1} , giving a transit time of $1.359 \mu\text{s}$; *ie* a change of transit time of 17 ns. Although precision ultrasonic test equipment could measure this small difference, most commercial test sets are not capable of the required accuracy. In addition, the temperature of the water bath is very important for accurate velocity measurements. A 1°C rise in the temperature, from 20 to 21°C , will increase the velocity of the compressional waves in water from 1483 to 1485.5 ms^{-1} , which for a 100 mm-long water path means a change in transit time of 113 ns. They therefore concluded that, for general inspection purposes, the use of attenuation measurements was preferable.

The ultrasonic attenuation was measured for the panels using three different pairs of transducers with measured centre frequencies of 2.5, 5 and 7 MHz in through-transmission geometry. The results were used to define a calibration curve for the initial resin system; see Figure 1. The measured attenuation of the signal was corrected for surface insertion losses before being converted to a value of the attenuation coefficient, α , in dB/mm. In the absence of a theoretical treatment two ways of fitting the data empirically were explored.

Initially a parabolic fit to the data was attempted using the form:

$$\alpha = n(f)V_v^2 \dots\dots\dots(1)$$

where n is a constant for a given frequency, f , and V_v is the void volume fraction. The constant n was found to be 0.251 for the 2.5 MHz data, 0.616 for the 5 MHz data and 0.912 for the 7 MHz data. The fitted curves using these three constants are also shown in Figure 1.

The dependence of n on frequency can be approximated from the above three values and the three theoretical curves may be expressed as:

$$\alpha = 0.0794 f^{1.27} V_v^2 \dots\dots\dots(2)$$

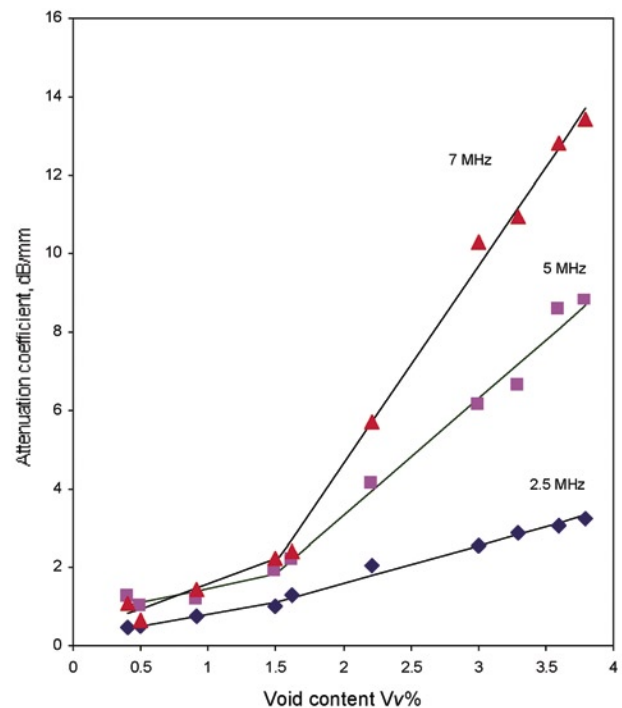


Figure 3. Variation of attenuation coefficient with void content^[6]. The curves show the fit to a bilinear form

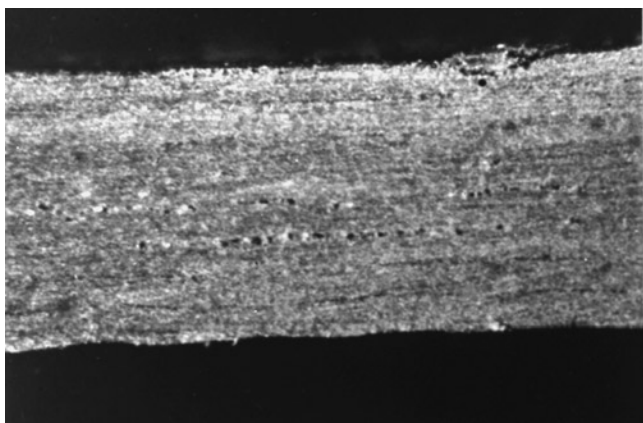
Figure 2 shows the curves generated using equation 2, compared to the experimental data.

A second fitting method took a bilinear form:

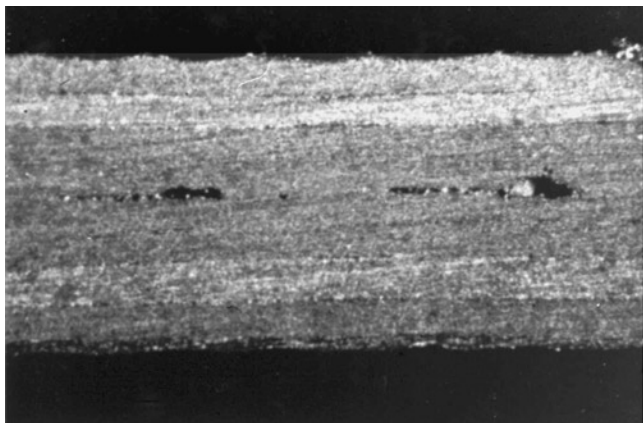
$$\alpha = a_1(f)V_v + b_1(f) \quad V_v < 1.5\% \text{ porosity} \dots\dots\dots(3)$$

$$\alpha = a_2(f)V_v + b_2(f) \quad V_v > 1.5\% \text{ porosity} \dots\dots\dots(4)$$

where a_1 , b_1 , a_2 , and b_2 are constants for a given frequency. This bilinear form (see Figure 3) was motivated by the observation, during destructive examination of the specimens, that there was a qualitative change in the character of the voids at about 1.5 % porosity. Up to 1.5 % porosity, the voids tended to be spherical, with the void diameter ranging from 5 to 20 μm . These voids were thought to be due to the various volatile elements present and there is some evidence that the size of the individual voids increases with V_v . Beyond about 1.5 % V_v , interlaminar voids caused by air entrapped between the laminates start to predominate. These were flattened and elongated and tended to be significantly larger than the volatile-induced voids; see Figure 4.



0.8 % Void volume fraction



2 % Void volume fraction

Figure 4. Typical void structure in carbon-fibre composite below 1.5 % V_v (mostly spherical voids) and above 1.5 % V_v (mostly flattened and elongated voids) as found by Stone and Clarke^[6]

Stone and Clarke concluded that the bilinear form appeared to provide a better fit to the experimental data.

The final part of the work by Stone and Clarke involved correlating the ultrasonic attenuation measurements with the interlaminar shear strength; see Figure 5. Previous studies^[2,3] had shown that the interlaminar shear strength was most seriously affected by the presence of porosity. The ultrasonic attenuation measurements were used instead of the porosity measurements as it was considered that the ultrasonic attenuation measurements were more reproducible than the porosity figures from the acid digestion method. For the fibre-resin system considered in this work, it can be seen that there was a good correlation between the ultrasonic attenuation and the interlaminar shear strength.

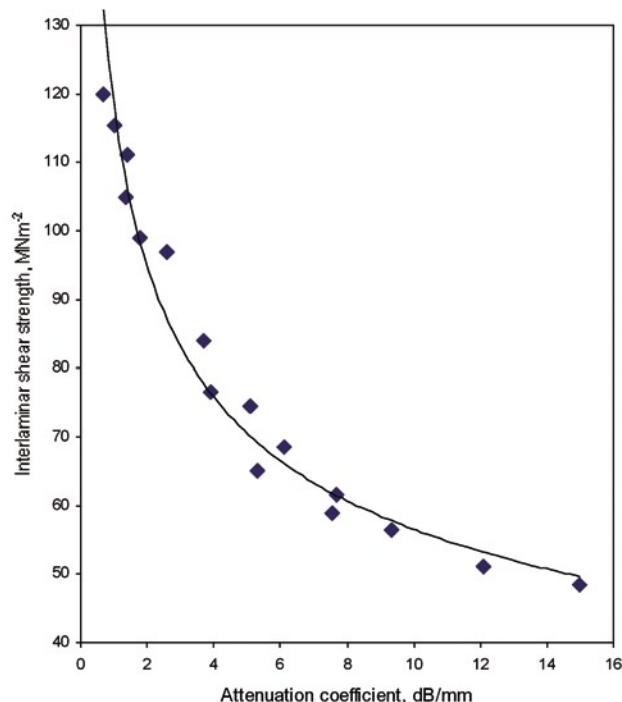


Figure 5. Relationship of interlaminar shear strength to attenuation coefficient for HTS fibres in an ERLA 4617/DDM matrix^[6]

Attenuation due to porosity arises from elastic scattering of the ultrasound due to the acoustic mismatch at the voids. As the mismatch in acoustic impedance between the matrix material and void is several orders of magnitude, it is reasonable to assume that the scattering will be dominated by the geometrical considerations alone and not by the material properties of the resin. In this approximation the increase in attenuation due to porosity would be similar for all resins, providing the morphology of the porosity does not change. It was therefore assumed that the data for the ERLA 4617 system could be read across to other resin systems.

A further programme of work was undertaken at RAE to check the validity of this assumption^[7]. The resin used for the follow-up study was DX 210 with the same volume fraction as the original study. The attenuation measured at 7 MHz centre frequency for

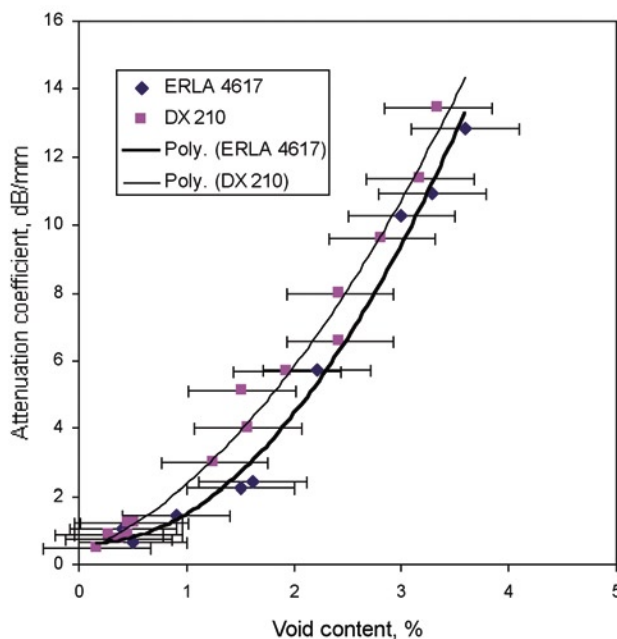


Figure 6. Variation in attenuation for two resin systems^[7]

the two resin systems is shown in Figure 6. It can be seen that the results are similar although the attenuation for the DX 210 system appears to be up to 1dB higher for intermediate void contents. The $\pm 0.5\%$ error bars for the acid digestion measurements have been added to the original Jones and Stone^[7] results and are based on accepted overall uncertainties in this method.

Martin^[8] presented a theoretical treatment of the relationship of the ultrasonic attenuation to the void diameter. In this analysis all the voids were assumed to be spherical. He compared his results to the experimental work by Stone and Clarke^[6]. Martin derived an expression relating the ultrasonic attenuation to the cube of the void radius, which was valid when the ultrasonic wavelength was much larger than the diameter of the void. So, if the void radius changed by a factor of two, the attenuation changed by a factor of eight. It was also shown that for measuring void content ultrasonically it was best to use the highest frequency possible that will not be totally attenuated for void contents in the range $\leq 4\%$ by volume. However, this is likely to move out of the regime where the ultrasonic wavelength is much larger than the diameter of the void.

Hsu and Nair^[9] related the slope of an attenuation versus frequency measurement to the void content. The work was based on a model of the voids, which assumed long cylindrical voids with an elliptical cross-section. This shape of void was chosen as it had been observed that in composite the voids tend to occur at the interface between the plies and are generally flattened and elongated along the axial direction of the adjacent fibres. For a set of seven unidirectional and quasi-isotropic laminates, reasonably good agreement was observed between the porosity measured ultrasonically from the attenuation slope and acid digestion measurements; see Table 1 and Figure 7.

Table 1. Comparison of porosity measured by attenuation slope and acid digestion^[9]

Sample	Porosity by attenuation slope, %	Porosity by acid digestion, %
1	0.5	0.2
2	0.2	0.32
3	0.8	1.14
4	1.1	1.25
5	1.8	2.04
6	2.5	2.82
7	3.4	4.05

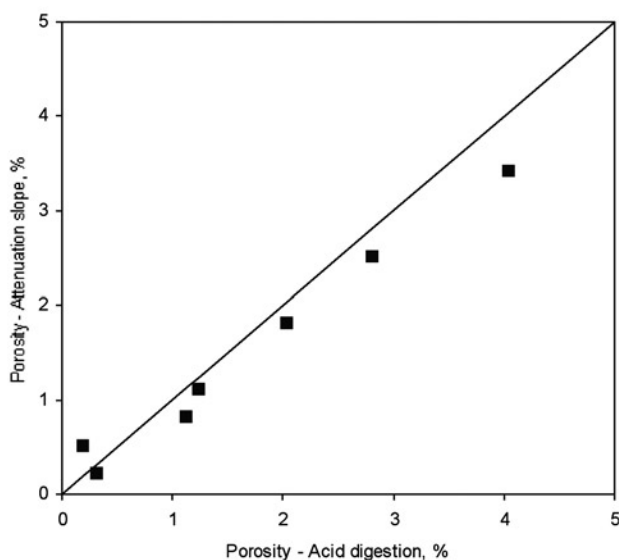


Figure 7. Comparison of porosity measured by attenuation slope and acid digestion^[9]. The line represents ultrasonic measurements exactly matching the acid digestion measurements and is shown for comparison with the experimental data

It was also noted by Hsu and Nair that porosity affects the spectral content of a broadband pulse. The shift in the centroid frequency when normalised by the centroid frequency of the void-free specimen showed a linear correlation with void content; see Figure 8.

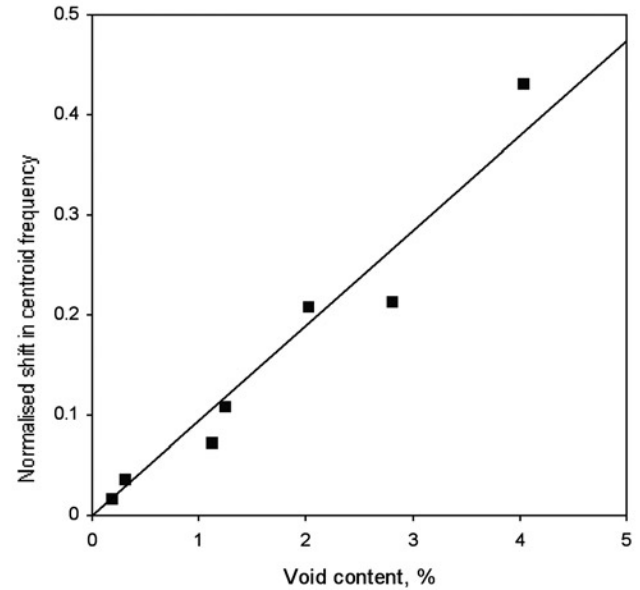


Figure 8. Correlation between void content and normalised centroid frequency shift for a 10 MHz transducer^[9]. The line is a linear best fit to the experimental data and has a slope of $0.095\%^{-1}$

The use of ultrasonic velocity measurements to determine both porosity and fibre volume fraction has been considered by other investigators. Reynolds and Wilkinson^[10] described a method to calculate the elastic constants of unidirectional fibre-reinforced materials containing matrix voids. This permitted the construction of calibration curves for given materials by means of which the measurement of two or more ultrasonic wave velocities may be converted to estimates of fibre volume fraction and porosity. Martin^[11] also developed a model relating ultrasonic velocities to both fibre volume fraction and porosity.

More recent work by Jeong and Hsu^[12] investigated the frequency dependence of the phase velocity (dispersion), which was ignored by both Reynolds and Wilkinson^[10] and Martin^[11]. When the wavelength of the ultrasound is of the same order of magnitude as the characteristic dimension of the voids it would be expected that over this frequency range the medium should show velocity dispersion. Dispersion effects are detected as frequency-dependent ultrasonic velocities. A pulse, being a superposition of many frequencies, will change its shape as it propagates through a dispersive medium. Jeong and Hsu undertook a theoretical analysis followed by an experimental study. The ultrasonic attenuation and dispersion are not independent according to Kramers-Kronig relations^[13]. Jeong and Hsu tested the relationship between the attenuation and dispersion using the theoretical local form of the Kramers-Kronig relation, which was found to hold experimentally for porous composite materials over the frequency range 2 to 10 MHz. The attenuation was found to be linearly dependent on frequency, with the attenuation slope ($d\alpha/df$) higher for laminates with higher void contents. The shape of the voids was found to have a great effect on the constant that relates void content to $d\alpha/df$. It was therefore important to use prior knowledge about the laminate structure and pore morphology to estimate the void content from the attenuation slope measurements.

The ultrasonic velocity decreased with increasing void content and the velocity dispersion increased as the frequency decreased. The velocity decrease was highest in unidirectional and quasi-

isotropic samples, lower in the woven carbon/epoxy samples and lowest in the woven carbon/polyimide samples.

Other ultrasonic techniques have also been used for porosity determination. Methods based on the detection of backscattered ultrasound have been widely deployed^[14-17]. These methods are, however, more difficult to adapt for routine in-service inspection purposes.

Summary of ultrasonic attenuation measurements for porosity

Measurement of the ultrasonic attenuation of composite laminates has been found in this review to be the most frequently used method to try to measure porosity. From the literature it is clear that several factors affect the relationship between the measured ultrasonic attenuation and the void content. It will be assumed that there is a general equation relating the measured ultrasonic attenuation, α , in dB/mm to the void content, V_v , of the form:

$$\alpha = k(f, d, s)C(V_v) \dots\dots\dots(5)$$

where k is dependent on the ultrasonic frequency, f , and the void shape, s and size, d . $C(V_v)$ is the function relating attenuation to V_v .

Stone and Clarke^[6] initially assumed:

$$C(V_v) = V_v^2 \dots\dots\dots(6)$$

and that k was a function of the ultrasonic frequency only, *ie*:

$$k = 0.0794f^{1.27} \dots\dots\dots(7)$$

However, Stone and Clarke concluded that a better fit was obtained using a bi-linear relationship of the form:

$$C(V_v) = a_1(f)V_v + b_1(f) \quad V_v < 1.5\% \dots\dots\dots(8)$$

$$C(V_v) = a_2(f)V_v + b_2(f) \quad V_v > 1.5\% \dots\dots\dots(9)$$

Jeong and Hsu^[12] found a linear relationship between the void content and the attenuation slope, $d\alpha/df$:

$$\frac{d\alpha}{df} = k(s)V_v \dots\dots\dots(10)$$

The parameter k was found to be dependent only on the void shape, s . This study was based on composite laminates manufactured from unidirectional, quasi-isotropic and woven laminates. The different material types produced different void shapes. The unidirectional and quasi-isotropic materials tended to have voids, which were flatter and longer. However, the voids in the woven laminates were more spherical in shape.

Other NDE methods for porosity measurement

Other methods for the determination of porosity have also been reported in the literature. Connolly^[18] reported the use of a thermographic method to measure the porosity of six carbon composite samples. The technique consisted of heating one face of the sample with a laser and observing the temperature rise on the other face as the heat diffused through the sample. A good correlation was obtained between the diffusivity and porosity for all six samples.

Gray *et al*^[19] investigated the use of microwaves for the estimation of porosity in polymer composites. This work is currently at an early stage and the reported results apply to air-filled microballoon inclusions in epoxy resin samples. They estimate that porosity changes of about 2 % should be detectable using the method. The technique reported should be applicable to glass fibre composite but is not a method easily adaptable for in-service use.

Conclusions

An in-service NDE method to determine the porosity of fibre-reinforced composite has not yet been found. Ultrasonic techniques based on attenuation and/or ultrasonic spectroscopy measurements appear to be the most promising areas for further investigation. Two factors need to be carefully considered. Firstly, the effects of the morphology of the voids on the porosity measurements require further investigation. Secondly, the method should ideally be independent of the fibre-resin system and volume fraction of fibre.

An experimental study is required to see if any of the methods presented in this review, or a combination of them, would allow for an independent porosity measurement in fibre-reinforced composite. Ideally, a method would be developed that could be embedded in a software-based NDE package and would provide percentage porosity from simple ultrasonic measurements that can be made in-service.

Acknowledgement

The authors would like to acknowledge the contribution of Dr David Bruce, Dstl, to the early stages of this review.

References

1. R A Garrett AGARD-CP-355, 'Effects of defects on aircraft composite structures,' p 19-1 to 19-34, 1983.
2. N C W Judd and W W Wright, 'Voids and their effects on the mechanical properties of composites – an appraisal,' SAMPE Journal, Jan/Feb, pp 10-14, 1978.
3. K M Uhl, B Lucht, H Jeong and D K Hsu, 'Mechanical strength degradation of graphite fibre reinforced thermoset composites due to porosity,' Review of Progress in QNDE, Edited by D O Thompson and D E Chimenti, 7B, pp 1075-1082, 1988.
4. D J Hagemmaier and R H Fassbender, 'Nondestructive testing of advanced composites,' Materials Evaluation, June 1979, pp 43-49.
5. J H Williams, H Nayeb-Hashemi and S S Lee, 'Ultrasonic attenuation and velocity in AS/3501-6 graphite fibre composite,' J of NDE, 1, No. 2, pp 137-148, 1980.
6. D E W Stone and B Clarke, 'Ultrasonic attenuation as a measure of void content in carbon-fibre reinforced plastics,' Non-Destructive Testing, June 1975, pp 137-145.
7. B R Jones and D E W Stone, 'Towards an ultrasonic attenuation technique to measure void content in carbon-fibre composites,' Non-Destructive Testing, April 1976, pp 71-79.
8. B G Martin, 'Ultrasonic attenuation due to voids in fibre-reinforced plastics,' NDT International, 9, pp 242-246, 1976.
9. D K Hsu and S M Nair, 'Evaluation of porosity in graphite epoxy composite by frequency dependence of ultrasonic attenuation,' Review of Progress in QNDE, Edited by D O Thompson and D E Chimenti, 6B, pp 1185-1193, 1987.
10. W N Reynolds and S J Wilkinson, 'The analysis of fibre-reinforced porous composite materials by the measurement of ultrasonic wave velocities,' Ultrasonics, 16, pp 159-163, 1978.
11. B G Martin, 'Ultrasonic wave propagation in fibre-reinforced solids containing voids,' J of Appl. Phys., 48, pp 3368- 3373, 1977.
12. H Jeong and D K Hsu, 'Experimental analysis of porosity-induced ultrasonic attenuation and velocity change in carbon composites,' Ultrasonics, 33, No 3, pp 195-203, 1995.
13. M O'Donnell, E T Jaynes and J G Miller, 'General relationships between ultrasonic attenuation and dispersion,' J Acoust. Soc. Am., 63, pp 1935-1937, 1978.
14. D E Yuhas, C L Vorres and R Roberts, 'Variation in ultrasonic backscatter attributed to porosity,' Review of Progress in QNDE, Edited by D O Thompson and D E Chimenti, 5B, pp 1275-1284, 1986.

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16. D Grolemond and C S Tsai, 'Statistical moments of backscattered ultrasound in porous fibre reinforced composites,' IEEE Trans on Ultrasonics, ferroelectrics and frequency control, 45, No 2, pp 295-304, 1998.
17. J Degrieck, N F Declercq and O Leroy, 'Ultrasonic polar scans as a possible means of non-destructive testing and characterisation of composite plates,' Insight, 45, pp 196-201, March 2003.
18. M P Connolly, 'The measurement of porosity in composite materials using infrared thermography,' J of Reinforced Plastics and Composites, 11, pp 1367-1375, December 1992.
19. S Gray, S Ganchev, N Qaddoumi, G Beauregard, D Radford and R Zoughi, ' Porosity level estimation in polymer composites using microwaves,' Mat. Eval. 53, No 3, pp 404-408, 1995.

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