FOLLOW-UP OF ENVIRONMENTAL DETERIORATION OF ADHESIVE JOINTS[1]

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CONTENTS

ABSTRACT

Samples of industrial structural adhesive joints were exposed to harsh environmental conditions. Periodical measurements allowed assessment of the amount of water absorbed by the samples, and destructive shear test

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demonstrated a constant reduction in the breaking-load of the joints. Ultrasonic echo signals from the samples were periodically recorded and subjected to signal processing. Features extracted from the echo-signals were found to be related to the reduction in the strength of the joints and/or the amount of the water absorbed by them. The empirical relations between the ultrasonic features and the mechanical properties of the joints implies the feasibility of using ultrasound to assess nondestructively, *in situ*, the condition of in-service structural adhesive joints.

1. INTRODUCTION

1.1. Structural Adhesive Joints

Adhesive bonding holds potential advantages over more traditional methods of joining structures and components such as welding and riveting. When properly applied, adhesives spread the load evenly over a large area, avoiding large stress concentration factors that could cause a premature structural failure. Adhesives are particularly useful when the linking of unlike materials is needed and when the adherends are made of fiber reinforced composites. Additional advantages of adhesive bonding include increased stiffness of the joint area, reduced machining requirements, a smoother external surface, sealing of the joint area and the prevention of galvanic corrosion when components made of different metals are linked together.

An essential requirement of a structural adhesive joint is that it maintains satisfactory long-term performance for the purpose for which it was designed. Joints are usually exposed to environmental conditions that affect both their strength and their long-term durability. The rate of adhesive joint degradation is determined by the joint characteristics, such as type of adhesive and pretreatment, cure cycle, etc, and the environmental conditions such as temperature, humidity, presence of chemicals, etc.

The most commonly encountered environmental factor is water. The presence of water in an adhesive joint can lead to the plasticization of the adhesive, the creation of microcracks, and the destabilization of the interfacial region between the adhesive and the adherend. The water-induced degradation is affected by the rate of moisture ingress and the inherent resistance of the joint to the presence of water. The latter is determined by the choice of adherend, pretreatment, primer and adhesive. The rate of water diffusion is affected by the relative ambient humidity, temperature and the presence of crazes and microcracks, especially in older specimens.

The empirically-based closed relationship between water diffusion and the degradation of mechanical properties of adhesive joints motivated a study into the monitoring of the degradation by means of feature-analysis applied to signals obtained from ultrasonic nondestructive testing, as described below.

1.2. Essentials of Ultrasonic Feature Analysis

In the application of ultrasonic nondestructive testing, low-energy vibrations are transmitted into a test object, where they are subjected to attenuation, scattering or reflection. The echo vibrations from the object are recorded and used to evaluate the condition of the specimen.

In the application of feature-based analysis, representative features are derived from the echo-signals, from both the time and frequency domain representations. It is desirable that the features should be chosen based on a profound understanding of the physical phenomena involved in the propagation of the ultrasonic vibrations in a given test-specimen, but features can be, and in many cases are, statistical parameters such as higher order moments of the signals.

The features are used to compare different classes of specimens, i.e., optimal and deteriorated adhesive joints, and to establish non-linear correlations between feature values and mechanical properties of the joints. In the application of the feature-based analysis extensive statistics, sophisticated mathematical algorithms and advanced signal processing techniques may be involved.

1.3. The Scope of the Study

The goal of the current study is to examine the feasibility of monitoring, by means of ultrasonic feature-based analysis, the degradation of structural composite adhesive joints exposed to harsh environmental conditions. Such monitoring is essential from the safety point of view, especially if the joints are used in the aeronautics industry, but it has an economic aspect as well, since it may prevent the periodic replacement of expensive parts which, in fact, may still be fit for use.

2. EXPERIMENTAL PROCEDURES

2.1. The Specimens

Composite adhesive joints of the single lap-shear configuration were used. The samples were 25.4 mm wide, with a 12.7 mm overlap between the adherends. The total thickness of the joint area was just over 4 mm, consisting of two adherends of thickness 2 mm each, and a bond thickness of approximately 0.15 mm.

Each laminated adherend was a 16 ply graphite/epoxy structure in a $[0_3, +45, -45, 0_3]_2$ sequence. The material used for the individual plies was a unidirectional prepreg, type 3502/As-4, manufactured by Hercules Aerospace. The adherends were fabricated from the individual plies in an autoclave process at 177°C and 700 kPA. A 100% nylon peel ply covered the surface of the adherend during its fabrication and was removed before commencement of the adhesive bonding sequence.

The surface pretreatment included Methyl-Ethyl-Ketone wiping, hand grinding with #180 followed by #280 grit papers, water rinse and drying. Bonding was accomplished with a film adhesive FM-300, 0.1 PSF, manufactured by American Cynamid. Curing of the adhesive was carried out at 177°C and 300 kPA.

2.2. Joint Strength

Destructive tests were carried out to determine the strength of the bonds as a function of time. The samples were tension tested to failure, parallel to the 0° direction of the adherends. Both loading grips were free to swivel so as not to induce external bending moments. The breaking loads and failure modes (adhesive or cohesive) were recorded.

2.3. The Ultrasonic Set-up

The ultrasonic experimental set-up consisted of a transducer, with a nominal central frequency of 1 MHz, a pulser/receiver, an oscilloscope, a digitizer, a computer and a X-Y scanning bridge.

2.4. The Experimental Protocol

The group of specimens consisted initially of 18 samples. three of them were destructively tested to assess the initial strength of the joints. The remaining samples were immersed in a tank of circulating tap water at 72°C together with a group of reference composite adherends. Periodically thereafter all the specimens were taken out of the bath to be weighed and subjected to an ultrasonic A-scan, using a pulse-echo mode with the ultrasonic beam at normal incidence to the specimens. Each digitized ultrasonic signal was averaged over 64 readings to improve the signal-to-noise ratio. One specimen was then destructively tested while the rest of the specimens were put back into the water until the next periodic examination.

3. RESULTS

3.1. Weight Gain of the Bonded Specimens

The mean increase in the weight of the bonded specimens due to the water uptake as a function of the time of exposure to the hot water is demonstrated in Figure 1. The figure reveals a monotonic increase in the weight of the bonded joints, which follows a typical Fickian pattern. The weight gain of the adhesive layers could be calculated from the overall weight of the specimens and the weight values obtained from the reference adherends.

3.2. Breaking-Load and Mode of Failure

The results of the destructive testing of the composite adhesive joints are presented in Figure 2, where the shear breaking loads are plotted as a function of the time of exposure to hot water. A monotonic decrease in the shear strength is revealed. For periods of time less than 4000h, a cohesive mode of failure was observed. For periods of time exceeding 4000h a mixed mode of failure was obtained: both cohesive and failure in the adherends fibers. For periods of time exceeding 6000h, a mixed mode of failure was obtained, which included cohesive failure, failure in the fibers and adhesive failure at the interface between the adhesive and adherends. One immediate result is that apparently, with a proper surface pretreatment to the adherends prior to bonding, it is the quality of the adhesive that determines the strength

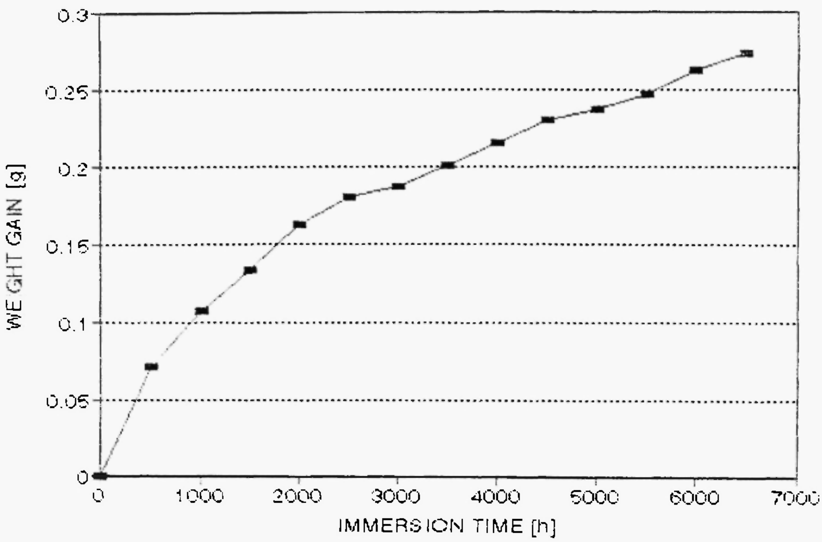

Fig. 1: The mean increase in the weight of the bonded specimens due to the water uptake as a function of the time of exposure to hot water.

of the joint. It is also noticeable that, for the configuration used in this study, the adhesive (interfacial) shear strength was less affected by the exposure to water than the composite adherends.

3.3. Ultrasonic Features Related to the Strength

A series of features was derived from each signal obtained from the ultrasonic testing. The features from all the signals were subjected to statistical and data processing techniques so as to indicate which of the features were the most sensitive to the deterioration of the joints. Two out of the series of features seemed to directly provide useful information as to the shear strength of the immersed joints. These two features were:

a. The variance of the power spectrum S(f), denoted f_1:

$$f_1 = \frac{M_2}{M_0} * \left(\frac{M_1}{M_2}\right)^2 \quad (1)$$

Where M_n is the n^{th} moment of the spectrum

$$M_n = \int S(f) f^n df \quad (2)$$

b. A parameter denoted f_2, based on the zero-crossing analysis of higher-order derivatives of the signals in the time domain. This parameter gives a measure of the 'distance' between a given and a reference signal. The signals are said to be different, with the difference being statistically significant, if the 'distance' is larger than a certain threshold. One can use any signal, but it is convenient to choose 'white noise' as the reference signal to which all other signals are compared.

In Figure 2, the mean values of f_1 are plotted as a function of the time of exposure to hot water, as well as the shear strength of the adhesive joints. Except for periods of time less than about 500h, the patterns are very similar (though on different scales), so that f_1 can serve as an indicator of the

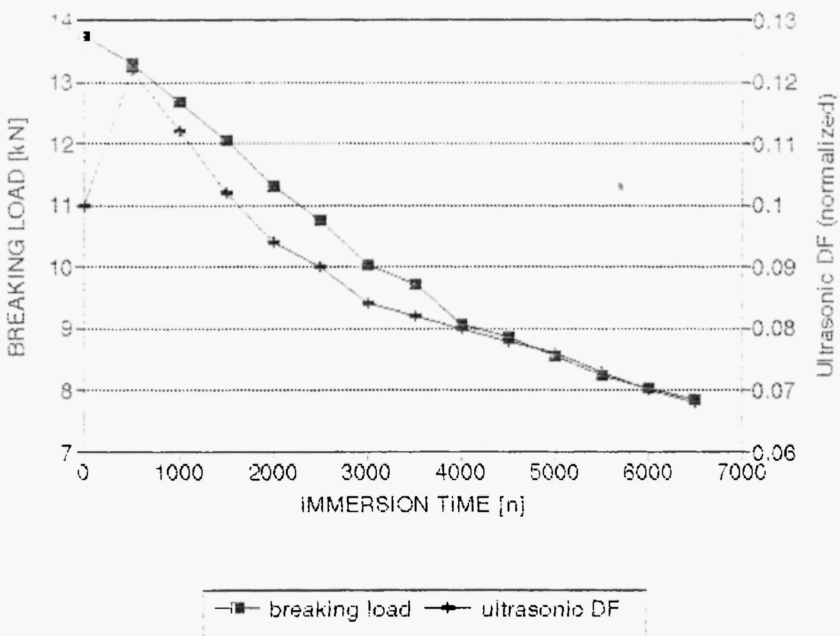

Fig. 2: The mean values of the ultrasonic parameter f_1, and the mean values of the shear strength of the adhesive joints, plotted against the time of exposure to hot water.

reduction in the shear strength of the joints. There is no satisfactory explanation for the different pattern obtained for time of exposure less than 500h.

In Figure 3, the mean values of f_2 are plotted as a function of the time of exposure to hot water, as well as the shear strength of the adhesive joints. The values of f_2 decrease with time up to about t=1500h, where there is a minimum, then increase up to t=4000h and from then on the values remain unchanged. A similar behavior was previously obtained for metal-to-metal joints.

The minimum value of f_2 is related to the condition when the ultrasonic beam starts to detect a separation between an adherend and the adhesive layer. The shape of the signal is then more and more dominated by the very well-defined modes of vibration of a plate, and the corresponding values of f_2 increase until they reach the value typical for a single plate.

The minimum value of f_2 corresponds to a 25% reduction in the shear

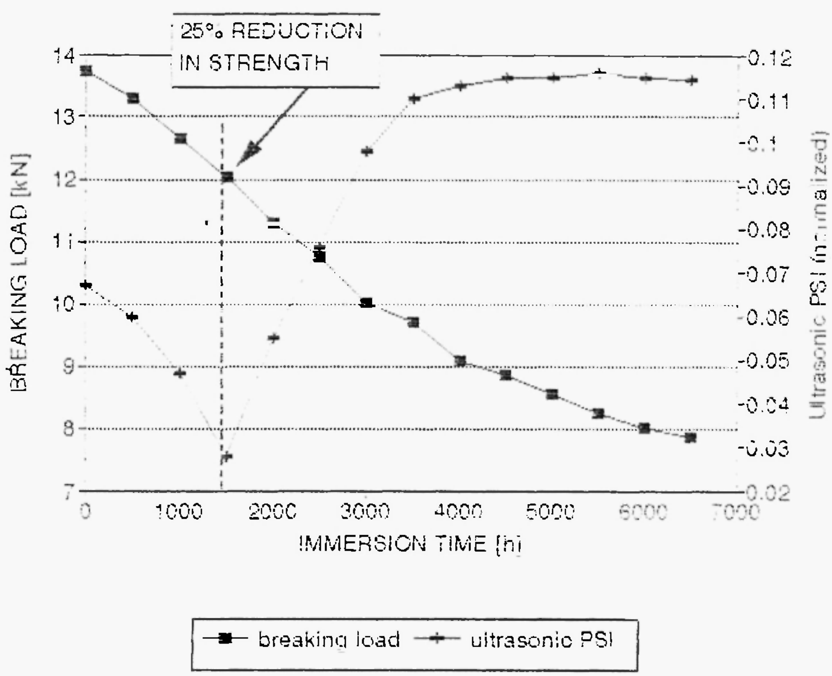

Fig. 3: The mean values of the ultrasonic parameter f_2, and the mean values of the shear strength of the adhesive joints, plotted against the time of exposure to hot water.

strength of the joints, and when reached, may serve as an alarm to indicate that the replacement of the compartment under inspection should be considered.

4. DISCUSSION AND CONCLUSIONS

The results obtained with the experimental set-up and procedures used in this study suggest the feasibility of monitoring the environmental deterioration of composite adhesive joints through changes in ultrasonic feature values. However, the fact that most of the features are based on statistical moments of the signals' presentation in either the time or frequency domains, and are somehow arbitrarily chosen, is scientifically disappointing. Unless features can be chosen based on a model of the mechanical and chemical phenomena that take place in the complex composite to composite adhesive joint system and their effect on the ultrasonic propagating beam, the search for sensitive ultrasonic features is mainly empirical.

Nevertheless, ultrasonic features may provide useful information as to the mechanical characteristics of composite joints. The relations between the ultrasonic features and the mechanical properties are not simple and require the use of predetermined calibration plots, based on the destructive testing of test-specimens of the same configuration and material as the object under evaluation. The preparation of such empirical calibration curves could be part of a quality assurance protocol designed to assess the integrity of joint structures manufactured in a mass-production line.

In this study, a very basic ultrasonic system was used, which is compact in size. Since the experimental procedure does not require high-level technicians, the system is suitable for *in situ* examination of in-service joints.

The results of this study may be improved if more complicated ultrasonic testing modes are used, such as oblique rather than normal incidence of the beam. The improved sensitivity that may be obtained using other modes should be evaluated against the increased complexity of the experimental procedures involved in those modes.

5. REFERENCES

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