ADHESIVE BOND TESTING BETWEEN COMPOSITE LAMINATES BY LASER SHOCKWAVE LOADING

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1 Introduction

Assembling materials by adhesive bonding has several advantages compared to other joining methods such as the use of fasteners or welding. Fasteners require drilling holes in the parts to be joined and both fastening and welding require significant investment in machinery. For metals, welded joints also generally produce a mechanically weaker heat affected zone. Adhesive bonding also has significant advantages for polymer-matrix composites. Drilling through composites has the drawback of cutting load-bearing fibers with adverse effects of possible delamination and excessive tool wear. It may also be economically advantageous to bond several small parts to make a large structure instead of having it co-cured. However, for all materials, the use of adhesive bonding for load-bearing structures is impeded by the absence of reliable nondestructive methods that can guarantee the strength of the joint, and in particular are able to very reliably identify the presence of near-zero strength “kissing bonds” [1].

Kissing bonds are undetectable by conventional ultrasonic inspection since the return echo from the interface in the pulse-echo technique does not depend upon the bond strength and only requires mechanical contact between the adherends. This is also the case for the transmitted echo. Although there have been many attempts to develop other ultrasonic approaches, such as using waves that propagate essentially along the bond line, none of these approaches has succeeded in detecting a weak bond other than those that are weakened by defects such as disbonds or porosity [1-3]. These defects can be detected by the well established ultrasonic inspection technique and in the case of porosity, also by x-ray radiography. Among possible causes of weak bonds are contamination of surfaces prior to bonding, inadequate surface preparation, degradation of the adhesive from improper storage, and inadequate mixing ratio for two-part adhesives. In all these cases, there can be good mechanical contact without defects, combined with poor mechanical strength, undetectable by established ultrasonic inspection techniques.

Ultrasonic techniques only apply weak stresses to the bond line and such weak stresses cannot reveal characteristics that are only apparent by applying significant stresses, like in destructive tests. Therefore, a reliable technique to identify such weak bonds requires application of a strong tensile stress across the bond line. A convenient approach that has been previously studied for evaluating the adhesion of coatings to their substrate and fibers to their matrix uses a pulsed laser to generate a large amplitude wave (shockwave) that propagates throughout the material [4-9]. This wave, being initially in compression, is converted by reflection on the back surface of the sample into a strong tensile wave that can pry apart the sample and disbond the assembly. This approach has been more recently extended to proof testing of adhesive bonds between carbon-epoxy laminates [10,11]. To probe bond strength, higher and higher tensile stress loading is applied by increasing the laser pulse energy step by step. A “good” joint will be unaffected under a given stress level whereas a weaker one will be damaged, allowing this method to evaluate the bond strength.

The principle of the method is described next in more detail. We then describe how the method is implemented, the instrumentation that has been developed and the fabrication of weak bond test specimens. Finally we present some results and indicate perspectives and future developments.
2 Principle of the approach

The approach is illustrated by the sketch shown in Fig.1. Real-time monitoring is provided by a laser interferometer that measures the back surface velocity. The signal from the interferometer allows the method to identify whether the bond has been broken or not. It also allows quantitative evaluation of the bond strength at the high tensile rate produced by the technique. For the sake of completeness, in the section below we reproduce the description that has appeared in publication [11].

We assume that the source size, which is about equal to the laser spot size, is several times the sample thickness. In this case the generated wave is essentially a compression wave propagating normal to the surface. Fig.2 shows the propagation distance-time diagram for the evolution of a compression wave generated at the top sample surface with a loading time duration $T$ and reflected by the free back surface as a tensile wave. In the triangle with baseline delimited by the times $t_1$ and $t_2$, the incoming compression wave is essentially compensated by the reflected tensile wave. If attenuation mechanisms are neglected, and for a typical shock pulse shape, the maximum tensile stress (represented as a white dot) begins at a distance from the back surface given by $DT/2$, and this tensile wave propagates unchanged until the next reflection. Here, $D$ is the shock wave propagation velocity which in the conditions used is essentially constant and close to the elastic wave propagation velocity $c$ since the pressure level is significantly below the Hugoniot Elastic Limit. Fig.2 therefore shows that there is a dead zone near the bottom of the sample. For the 4-ply quasi-isotropic composite schematically represented in Fig.2, this zone extends to the top of the last ply assuming a loading duration of about 100 ns and a ply thickness of about 150 $\mu$m ($D \approx 3000$ m/s).

The pressure $P(z,t)$ at any instant $t$ and at a depth $z$ inside the plate is equal to the sum of the pressures produced by the wave propagating in the positive $z$ direction and the one propagating in the negative $z$ direction:

$$P(z,t) = \rho \left[ D \cdot u^+(z,t) + (-D) \cdot u^-(z,t) \right]$$  \hspace{1cm} (1)

where $u^+(z,t)$ and $u^-(z,t)$ are the particle velocities of waves propagating toward positive and negative $z$ respectively. The minus sign comes from wave propagation in the negative $z$ direction. The $z$ axis origin is taken to be the back surface where velocity measurements are made and $z$ is oriented positive from the front loading surface toward the back surface. Under the assumptions of one-dimensional propagation in a homogeneous material and no attenuation, at the free back surface and after the propagation of $u^+(z,t)$ and the retro propagation of $u^-(z,t)$ this equation becomes:

$$P(z,t) = \frac{1}{2} \rho D \left[ u(0,t+|z/D|) - u(0,t-|z/D|) \right]$$ \hspace{1cm} (2)

The factor $1/2$ comes from the total reflection $u^+(0,t)$ = $u^-(0,t)$ of the wave at the free surface: $u(0,t) = u^+(0,t) + u^-(0,t)$. The last equation gives the pressure at any depth, at any time during the experiment. The rupture threshold is then given by the following equation, which applies for loading just below the rupture threshold:

$$P_{rupt} = \frac{1}{2} \rho D \left[ u(0,t') - u(0,t' - 2z_{rupt}/D) \right]$$ \hspace{1cm} (3)

In this equation, $t'$ is the time at which the damage is identified on the velocity signal and $z_{rupt}$ the distance of the damage from the back surface. $t'$ appears on the back surface with a delay of $|z_{rupt}/D|$ from the time $t'_{rupt}$ at which rupture occurs.

As shown by experiments, the velocity signals obtained with all laser loadings below rupture are all essentially superimposed after normalization. This is shown in Fig. 3 for a laminate without a bond, reproduced from a previous publication [11]. This is in agreement with numerical simulations performed with LS-Dyna as shown in Fig. 4. This means that for all loadings below the rupture threshold, propagation is in the elastic regime, no plastic deformation occurs, and non-linear effects are very weak. A difference only occurs for loading cases above the disbond threshold. In this case the tensile wave is reflected by the opened interface as a compression wave, which gives in turn a positive increase in velocity measured on the back surface, i.e. an increase of the out-of-plane motion of this surface. The time at which this difference is first
identified defines $t'$. The depth of the rupture $z_{rupt}$ is better measured by post shock ultrasonic or laser-ultrasonic inspection. In the case of very strong bonds or weak materials, it is also possible that the parent material fails before the bond. Fig. 5 is a schematic representation of the procedure to identify $t'$ and to evaluate $P_{rupt}$. $P_{rupt}$ is calculated from the signal recorded just below rupture: we note that $1/2 \rho D \ u(0,t')$ corresponds to the stress applied to the joint by the incoming compression wave whereas $-1/2 \rho D \ u(0,t')-2 \ z_{rupt} / D)$ is associated with the reflected tensile wave.

3 Implementation

3.1 Shock wave generation

To produce a strong compression wave, laser pulse generation in the ablation regime with plasma confinement is used. To maximize the pressure in the sample, a confining material with large acoustic impedance is required. This material has to be transparent to the laser light. Although confinement with a glass window produces a strong shockwave, the window is broken by each laser shot, so confinement with water is used, as shown in Fig. 6. Since it would be unacceptable in practice to damage the surface of the material, common black electrical tape is stuck to the surface. A Q-switched Nd:YAG laser operating at 1.06 µm and delivering a pulse of about 8 ns duration and 2 J energy has been used so far. As we have previously shown, by measuring the velocity at the back of a transparent block of epoxy subjected to the same loading configuration as shown in Fig.6, the protective tape has the effect of increasing the duration of loading [11]. This duration is on the order of 100ns instead of about twice the laser pulse duration that could be expected [12]. Since high frequencies are cut-off in composites by damping and scattering mechanisms, a longer loading duration is beneficial for maintaining the tensile stress level over a longer propagation distance.

3.2 Real time monitoring

As we have seen, the stress applied throughout the sample can be determined from measurement of back surface velocity. This velocity is determined from the Doppler frequency shift produced on a light beam reflected by the surface. The light frequency change $\Delta v$ is proportional to the change of velocity $\Delta u$ as follows:

$$\Delta v = 2\Delta u/\lambda,$$

where $\lambda$ is the laser wavelength, $\lambda$ being 1.06 µm, a change of velocity of 100 m/s gives a frequency shift of about 100MHz. This frequency shift is then tracked by the interferometer. Although for shock experiments time-delay two-wave interferometers known as VISAR are traditionally used as velocimeters [13], we have developed one that is intrinsically simpler and makes use of the detection laser used for post-shock validation. It uses a solid Fabry-Perot etalon [14]. Its principle of operation is described next.

The principle is similar to the use of a Fabry-Perot interferometer in optical spectroscopy. In spectroscopy, spectral information on the light source is obtained using a circular aperture to select part of the central fringe pattern in the focal plane of a lens and the spectrum of the source is obtained by scanning the path difference (i.e. the Fabry-Perot optical thickness). Here, the frequency of the source is actually changing with time due to the Doppler effect produced by the surface motion and the Fabry-Perot has a fixed thickness (Fabry-Perot etalon or FPE). The signal related to surface motion appears on the detector behind the aperture at the center of the fringe pattern: see Fig. 7.

To use the Fabry-Perot etalon as a velocimeter, the frequency of the probing laser is set somewhere along the slope of a peak. Since by the Doppler effect, as mentioned above, a change of frequency proportional to the surface velocity is produced, this results in an output from the Fabry-Perot detector of an intensity signal indicative of the surface velocity; see Fig. 8, which illustrates the principle. The output signal is only proportional to velocity if the linear portion of a peak slope is used. If this is not the case, the response has to be calibrated by recording the peak shape versus frequency. Actually, the peak shape is dependent upon the opening of the aperture after the FPE, i.e. it depends of the angular tilt of the optical waves propagating inside the FPE. Having a very limited angular range around zero, i.e. a very small aperture yields poor sensitivity since little light is collected, so the diaphragm aperture has to be
opened to some extent. The effect of aperture size has been modelled and the results are shown in Fig. 9. The narrowest peak is only obtained for a tightly closed diaphragm. This has also been verified experimentally by sweeping the frequency of the detection laser: see Fig. 10. The final choice of the response curve of the FPE is shown in Fig. 11: it provides optimum sensitivity and a dynamic velocity range of about 200m/s, adequate for all the loadings used.

To get a reliable calibration curve that allowed translation of the recorded signal to a velocity signal, the voltage which controls the frequency sweep had to be calibrated in terms of frequency change. This was done by using an optical Michelson-type fiber interferometer with arms of very different lengths. By counting the fringes that appear during frequency sweeping, precise calibration is obtained. The final calibration curve is shown in Fig. 12: it relates the normalized signal (signal measured by the detector after the FPE divided by a signal measured by another detector before the FPE) to the change in velocity. To convert from frequency change to velocity change Eq. 4 has been used.

As shown in Fig. 12 by a large dot, after calibration by frequency sweeping, an initial frequency set point was chosen. This set point was chosen to provide good sensitivity to surface velocity, as well as ensuring a proper dynamic range for the velocities encountered in the experiments. Also, since a given signal amplitude can be associated with two frequency shifts or velocities, it is important to avoid going over the peak of the response for the maximum velocity to be measured in order to avoid ambiguity. The usable range of velocity change is shown in Fig. 12 by a heavy line. The laser frequency was then locked to the set point with a feedback loop acting on the frequency controller for the laser. This ensured that thermal drifts of the laser frequency or of the FPE resonance frequency are automatically compensated.

The FPE is mounted with its associated optics in a rigid structure that is itself located in a rack. The detection beam, which comes from a long-pulse single frequency Nd-YAG laser is sent onto the back surface of the sample using an optical fiber. Light reflected by the surface that carries information on its velocity during shockwave loading is also fiber-coupled to the FPE unit.

### 3.3 Post-shock validation

Post-shock validation by ultrasonic inspection is necessary to validate any indications of rupture given by the real-time velocimeter, since this indication is often difficult to identify when loading is just above threshold. Ultrasonic inspection using the pulse-echo time of arrival technique provides the location of the disbond, which is necessary to evaluate bond strength, as seen above. We have performed this inspection with a laser-ultrasonic system that uses the same kind of detection laser used for the velocimeter [15]. The generation laser is a Transversally Excited Atmospheric Pressure CO$_2$ laser (TEA-CO$_2$), as generally used for composite inspection. This laser has a wavelength within the absorption bands of the surface resin, epoxy or another polymer matrix, and from the through-depth distributed absorption, ensures efficient generation without damage to the material. This operation can be done from the loading surface after the protective tape is removed (generation over the tape would produce longer and weaker echoes compromising resolution of the echoes from interfaces) or from the back surface. Although in this work the sample has been analyzed with a laser-ultrasonic inspection system separate from the laser shock system, shockwave loading, velocimetry and post-shock validation could be combined into a single system since the shock, its real time monitoring and post-shock validation could use the same detection laser. This has actually been implemented for a different bond testing application and is planned to be implemented for composites.

### 4 Test samples

The technique was first demonstrated with bonded quasi-isotropic carbon-epoxy laminates prepared with 2 areas of different adhesion strength. One area of each laminate was simply solvent cleaned, whereas the adjacent area was corona discharge activated to promote higher adhesion strength. Hysol EA9394 adhesive was used. As expected, very different adhesion strengths were measured, typically 150 MPa and 340 MPa [11]. It should be noted that these values were measured at very high strain-rates and are higher than typical quasi-static values. Velocimeter indications of disbonding were
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found to correlate well with post-shock laser-ultrasonic inspection.
To develop a correlation between the values measured by the laser shockwave technique and the usual quasi-static values, there was a need to make test samples with a distribution of adhesion strengths, having for example values equal to nearly zero (which may be identified as a kissing bond), 25%, 50%, 75% and 100% of the optimum strength. To make these controlled weak bonds, two approaches have been used [16].
In the first approach, one composite adherend was metalized with a very thin layer of titanium (less than 100 nm) and a custom bond primer made from a mixture of silanes (γ-GPS and PTMO) was applied. Bond strength was tuned by varying the ratio of the concentrations of γ-GPS and PTMO. The bond with the other adherend which was not metalized was made full strength. For this approach, Cytec FM73 and Hysol EA9394 were the adhesives used.

In the second approach, the mix ratio of a two-part adhesive was modified by reducing the hardener fraction. This modification caused weakening of bond strength but had the drawback of inducing very long cure times and/or an unstable cure state (i.e. residual reactivity). To avoid this problem, a predetermined amount of a third chemical component (cyclohexylamine) was added to the epoxy resin and its hardener to rebalance the reactive fractions. For this approach, two-part Hysol EA9394 was the adhesive used.
The fabricated test specimens were destructively tested by double cantilever beam (DCB), i.e. Mode 1 through-thickness tension testing [17]. Although both approaches were demonstrated to be sound and able to create controlled weak bonds, it proved difficult to consistently obtain specific strength values for the lower strength bonds and significant variability was observed between samples made under the same conditions. At the present time, the desired range of distributed adhesion strengths going from nearly zero to 100% in steps of 25% has not been repeatably demonstrated and further work is required.

5 Results

We present below example results obtained on a test specimen bonded with EA9394 and the customized surface treatment. The bond made to the metalized adherend was intended to have 25% strength, but turned out to have about 64% of the maximum strength with a variance of about 14% between different samples (as tested by DCB). The other adherend was not modified and had 100% bond strength. Prior to laser shockwave loading, all the samples were inspected by digital x-ray radiography to check for porosity in the composite or bond line and by laser-ultrasonics to check for porosity and pre-existing delamination or disbond. This prior inspection ensured the laser shockwave testing could be performed over a defect-free area having only weak adhesion properties.

Fig. 13 shows the back surface velocimeter signals obtained from this test specimen after normalisation of the signals for different loadings. As described above, disbonding was identified by the appearance of a significant offset after signal normalization. This occurred clearly at about 1.2 J energy loading. Fig. 14 shows the confirmation of disbonding by laser-ultrasonic inspection. Using the approach described in Fig. 5 and a formula similar to Eq. 3 (but more accurate, to take into account the transmission of the stress through the 4-ply laminate as derived in [11]), the bond strength of this sample was evaluated to be about 80 MPa in the high strain rate regime of the experiment: see Fig. 15.

6 Conclusion and perspectives

The laser shockwave technique has been shown to be able to detect weak bonds between laminates. The developed technique is non-invasive if the bond is sufficiently strong for the shockwave loading applied. However, a correlation between the value measured by the technique at high strain rate and the strength measured by established destructive testing techniques, such as DCB, remains to be established. To reach this aim, future efforts should be focused in two directions. In the first, one should try to minimize the variability of weak bonds produced by one or both of the approaches described here. The other direction is to extend this work using a much more energetic shockwave loading laser (10 J or more) with a long pulse duration (300 ns and more). With such a laser, the dead zone shown in Fig. 2 will extend much further from the back wall of the part. The stress loading being essentially low
frequency will propagate deep inside the material without significant attenuation, thus proof-loading the bond to the same stress level as the laminate. Long pulse duration will avoid causing a failure of the composite before the bond in all cases in which the bond is weaker than the composite. Higher energy will also allow the use of a larger test laser spot, which is important for testing assemblies thicker than the 4 or 8-ply used in this work. This is necessary because a small spot causes diffraction effects and stress loading which is significantly different from a plane compressive wave, with complicated tensile and compression zones throughout the thickness of the material.

In the shorter term, since not all the fabricated weak bond samples have been tested, we will continue laser shockwave testing on these to collect additional data.

Fig. 1. Principle of the laser shockwave technique for probing an adhesive joint. The wave which is initially in compression is reflected by the free back surface as a tensile wave that performs tensile proof testing of the joint.

Fig. 2. Time-space diagram of the propagation of a shock wave pulse with duration $T$ and schematic representation of the cross-section of a quasi-isotropic 4-ply composite. Dark and light gray areas represent respectively the compression wave (pressure higher than the average pressure) and the tensile wave (pressure lower than the average pressure). The arrow indicates the depth where the maximum tensile stress is applied, corresponding in this example to the top of the bottom ply.

Fig. 3. Back surface velocity signals measured under 1200 mJ (grey curve) and 800 mJ (black curve) laser shock pulse energy on a 4-ply laminate.
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Fig. 4. Back surface velocity signals measured (black and noisy curve) and simulated (grey curve) for a 400mJ laser pulse in a 4-ply laminate.

Fig. 5. Schematic representation of velocity signals from the back surface. The black and grey signals represent respectively a signal above and slightly below damage threshold. The black signal has been normalized to the grey signal maximum.

Fig. 6. Experimental setup for strong compression wave generation ensuring protection of the sample surface.

Fig. 7. Principle of the Fabry-Perot interferometer as used in optical spectroscopy. A series of rings can be observed in the focal plane of the lens if the source gives a sufficient angular range. The detector (not represented here) is located behind an aperture centred on the rings so as to receive only part of the central fringe.
Fig. 8. Frequency demodulation with a Fabry-Perot. The optical frequency of the probing laser is tuned to somewhere along a resonance peak to provide demodulation.

Fig. 9. Theoretical resonance curves of the FPE for different diaphragm openings. The narrowest curve is obtained for the smallest opening; the curve broadens with the opening diameter.

Fig. 10. Experimental resonance peaks of the FPE for different diaphragm openings.

Fig. 11. Effect of a slight tilt of the FPE with respect to the axis defined by the diaphragm aperture: the broadest curve is obtained for optimum centering whereas the curve with the higher peak is obtained after slight tilt. This is the response chosen for the velocimeter: it gives dynamic ranges (10-90%) of about 200 and 550 m/s, for the rising and falling slopes respectively.
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Fig. 12. Calibration curve of the velocimeter. The change of velocity at any time can be deduced from the measured normalized signal by using this curve. The dot indicates the initial set point.

Fig. 13. Velocimeter signals obtained for a bonded 4-ply over 4-ply stack for laser energy loading clearly above (1.2 J), below (0.8 J) and at about (1 and 1.12 J) the damage threshold. The various signals have been normalized and the time of indication of rupture is shown by a vertical line.

Fig. 14. Laser-ultrasonic C-scans for various laser energies. Disbonding starts to be detected at about 1000mJ - 1120mJ energy loadings.

Fig. 15 Determination of the bond strength from the velocity signal just below threshold according to the method indicated in Fig. 5.
References


