Laser spallation: A novel technique to evaluate thin film interface strength

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Abstract

The versatility of laser spallation technique in evaluating dynamic interface strength of a range of dissimilar bi-materials is demonstrated. Four different combinations, metal/polymer, metal/composite, ceramic/polymer and ceramic/metal, are considered in this study. Interfacial failure is instigated by laser induced stress waves which load the materials at a strain rate of ~ 10^7/s. Failure initiation is identified by optically analyzing the free surface of thin films. Profilometric scan across the spallation zones confirms interfacial failure in all specimens. Michelson interferometer in combination with laser spallation technique is employed to perform in-situ free surface displacement measurements in calibration samples. The interferometric data along with the wave propagation analysis is used to obtain substrate stress histories which in-turn utilized in computational models to infer the bilayer interface strength. The dynamic interface strength of aluminum/epoxy, fused quartz/epoxy and fused quartz/copper is measured to be, 264 MPa, 205 MPa and 146 MPa, respectively.

Keywords: Laser spallation, bi-material interface, Michelson's interferometer, dynamic interface strength, stress wave.

1. INTRODUCTION

The layered materials system has wide variety of engineering applications ranging from in micro-electronics components to aerospace industries. Often multilayer structure is developed by consecutive deposition of thin layers such as in semiconductor devices or by laying down several layers simultaneously as in case of laminated composites. The reliability and functionality of such systems inevitably depend upon the strength of the layered interfaces. The failure behaviour further complicates when the interfaces are subjected to high strain rate loadings.

Often conventional techniques such as peel [1,2], scratch [3], stud-pull [4], bulge [5] and four point bend [6,7] tests are used to characterize quasi-static bi-material adhesion strength. Sometimes split Hopkinson's bar test is employed to evaluate dynamic adhesion characteristics of the interfaces [8,9]. Although the test methods are well established for conducting experiments at macro scale, the methodologies failed to evolve for the cases when the material dimensions are constrained to micro and nano levels. The laser spallation

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technique [11–16] provides an alternative where a laser-generated high amplitude acoustic stress pulse is employed to dynamically load the film interface in non-contacting mode. The method was first introduced by Yang [10] and Vossen [11]. Gupta and co-workers [12,13] further developed the technique and coupled it with Michelson's interferometry to measure thin film interface strength. The method had been used to characterize metallic, ceramic and polymer film interfaces where the film thickness ranged from few hundred nanometers to few millimeters [12-16].

In this investigation, the versatility of non-contact laser spallation technique to characterize the adhesion strength of various dissimilar interfaces is demonstrated. Various combinations of test specimens are prepared with two different substrates, fused quartz (FQ) and aluminum (Al). The epoxy, copper and single ply of unidirectional carbon lamina are used as the film material. Failure at bi-layer interface is instigated by laser induced stress pulses and in-situ quantitative evaluation of interface strength is performed by employing Michelson interferometry. Optical micrographs in combination with profilometric scan confirm the interfacial failure of test films.

2. MATERIAL AND SAMPLE PREPARATION

Aluminum substrates are prepared by milling the bars of Al-6061-T6 to 750 µm thicknesses, followed by cutting them into 25 mm x 25 mm square samples. These plates are polished by using 5 µm and 1 µm alumina slurry to achieve a good surface finish. The fused quartz (FQ) plates of dimension, 25 mm x 25 mm x 1.3 mm, are used as another substrate material. The test samples are fabricated next by depositing epoxy film on top of the substrate. The epoxy system is prepared by mixing Diglycidyl Ether of Bisphenol-A (DGEBA) epoxy resin and Tri-ethyl tetra amine (TETA) hardener in 10:1 weight ratio. Few drops of the mixture are placed onto the substrate. This is followed by adjusting the cellophane tape wrapped glass slab and a pre-calibrated dead weight on top of it to achieve the desired film thickness. The entire assembly is left at room temperature for about 15 hours for curing. The glass slab is carefully removed and the substrate/film assembly is kept for about 15 days to ensure complete curing. When copper is used as a test film, thermal evaporation is employed for deposition. For preparing Al/composite sample, 25 mm x 25 mm square shaped (unidirectional) carbon prepreg is cut and laid down onto the aluminum substrate. The assembly is compressed in a mold and cured at 120°C for 4 hours, followed by maintaining the temperature of the oven at 180°C for the next 8 hours. The samples were left at room temperature in humidity controlled environment for a period of 15 days. In current
investigation the thickness of the copper, epoxy and composite films are considered to be 0.5 µm, 15 µm and 125 µm, respectively. Prior to conducting experiments a thin sacrificial layer of aluminum is deposited at the back surface of the substrate which is further spin coated by a 10 µm water glass layer. In addition the calibration samples are also prepared by depositing 100 nm Al layer on top of the Al and FQ substrate (instead of the test films). The back surface of the sample was prepared with energy absorbing and confining layers as discussed earlier.

3. EXPERIMENTAL DETAILS

The schematic representation of the laser spallation setup in combination with Michelson's interferometry is shown in Fig. 1. A Q-switched Nd:YAG laser of 5 ns pulse width with varying energy content (0-300 mJ) is impinged on the rear surface of the substrate. Localized ablation of Al film develops compressive stress pulse in the substrate which propagates across the interface, reflects as a mode converted tensile pulse from the free surface of the film and loads the interface in tensile mode. The magnitude of the laser fluence is gradually increased till the interfacial failure is initiated. The strain rate associated with the experiment is ~10^7/s.

Free surface displacement history in calibration sample is recorded by employing Michelson’s interferometry as illustrated in Fig. 1. The optical path difference between the reference and the probe beam develops interference fringes. The variations in the fringe intensities are captured using photo-diode and an ultra high frequency oscilloscope. The fringe intensity \(I(t)\) is associated with the fringe order \(N(t)\) by the following equation.

\[
I(t) = \frac{I_{max} + I_{min}}{2} + \frac{I_{max} - I_{min}}{2} \sin 2\pi N(t),
\]

where \(I_{max}\) and \(I_{min}\) are the maximum and the minimum intensity of the fringe in consideration, respectively. This is followed by obtaining out-of-plane displacement \(u(t)\) by employing Doppler shift equation[17].

![Figure-1: Laser spallation setup with Michelson's interferometry](image-url)
\[ u(t) = \frac{\lambda}{2} N(t). \]  

(2)

In the above equation \( \lambda \) (= 514 nm) represents the wavelength of the probe beam. Next, the substrate stress history \( \sigma_{\text{sub}}(t) \) and interface stress history \( \sigma_{\text{int}}(t) \) are evaluated by [14],

\[ \sigma_{\text{sub}}(t) = -\frac{1}{2}(\rho C_d)_{\text{sub}} \frac{\partial u}{\partial t}, \]  

and

\[ \sigma_{\text{int}}(t) = (\rho h)_{\text{film}} \frac{\partial^2 u}{\partial t^2}, \]  

respectively. In the equations, \( \rho, h \) and \( C_d \), represent the density, thickness and dilatational wave speed, respectively.

4. CALIBRATION PROTOCOL AND REPRESENTATIVE DATA

While in-situ measurements are performed on Cu film specimens (by applying Eqs (1-4)), hybrid experimental/numerical approach [15,16] is adopted for evaluating interface strength in case of epoxy and composite film specimens due to optically non-reflective films. In the later interferometric measurements are performed on calibration samples. The substrate stress histories correspond to various laser fluences are obtained by applying Eqs (1-3) on fringe intensity data. The substrate stress is next used as transient loading in planar wave propagation simulation to evaluate the associated interface stress history. Once the calibration chart correlating laser fluence with the interface stress peak is prepared, the laser spallation experiments are performed on actual test specimens. The laser fluence correspond to the failure initiation is noted and the associated stress from the calibration chart is inferred as the interface strength.
Table 1. Elastic properties of material used in computational wave propagation analyses.

<table>
<thead>
<tr>
<th>Material</th>
<th>Density ‘ρ’ (kg/m³)</th>
<th>Elastic modulus ‘E_d’ (GPa)</th>
<th>Poisson’s ratio ‘ν_d’</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy</td>
<td>1195</td>
<td>4.838</td>
<td>0.36</td>
</tr>
<tr>
<td>Fused quartz (FQ)</td>
<td>2200</td>
<td>72.93</td>
<td>0.168</td>
</tr>
<tr>
<td>Copper (Cu)</td>
<td>8924</td>
<td>130.07</td>
<td>0.3424</td>
</tr>
<tr>
<td>Aluminum (Al)</td>
<td>2700</td>
<td>69.95</td>
<td>0.36</td>
</tr>
</tbody>
</table>

The two dimensional bi-material geometry, modeled in ABAQUS/CAE-6.12, is shown in Fig. 2. Four noded plane strain element CPE4R is used to mesh the geometry with the smallest element size of 0.00012 mm. The material properties, given in Table 1, are assigned to the respective layers. Roller supports are used at the top and bottom surfaces of the substrate to ensure planar wave propagation. The substrate stress history obtained from the calibration experiments is applied to the back surface of the substrate without embedding energy dissipation mechanisms in the simulations.

The representative interferometric fringes shown in Fig. 3 (a), correspond to the calibration experiment performed on Al substrate at 210 mJ/mm² laser fluence. The associated displacement history (Fig. 3(b)) evaluated by using Eq. (2) shows a monotonic increase till 197 ns (turn-around point), which is followed by a decreasing displacement trend. Figure 3(c) illustrates the corresponding substrate stress history, calculated by applying Eq. (3), which shows a peak value of ~ 500 MPa. The associated interface stress history obtained from computational analysis is next illustrated in Fig 3(d) with the maximum value of the tensile stress of ~ 265 MPa.

![Figure 3](image-url)
5. RESULTS AND DISCUSSIONS

The optical micrographs illustrating interfacial failure in tested specimens is shown in Fig. 4. The failure has initiated at Al/epoxy interface at 210 mJ/mm$^2$ laser fluence (see, Fig. 4(a)). At higher laser fluence the film got completely spalled off. Corresponding image and the profilometric scan across the spalled zone are shown in Figs. 4(b) and (b’), respectively. The surface scan data confirms interfacial failure in 15 μm epoxy film. Failure images in Figs. 4(c) and (d) correspond to the FQ/epoxy specimens. At 65 mJ/mm$^2$ laser fluence the film just got delaminated from the substrate whereas significantly larger spallation spot is evident from Fig. 4(d) at higher laser fluence. Figure 4(d’) again confirms interfacial failure. The darker shadow around the spallation zone is the region of laser incidence the FQ substrate.

![Image](image_url)

**Fig. 4.** Spallation spots at interfacial failure initiation and at higher laser incidence along with the profilometric scans for the later case. The subfigures correspond to the test samples: (a-b’) Al/epoxy, (c-d’) FQ/epoxy, (e-f’) FQ/Cu, and (g,h) Al/Carbon ply composite.
Unlike in epoxy film cases, the optical micrograph shows a few spallation spots at interfacial failure initiation in FQ/Cu specimen (see, Fig. 4(e)). While conducting experiments the spin coated water glass thickness was reduced from 10 \( \mu m \) to 4 \( \mu m \) for optimizing the laser fluence at failure initiation. Figures 4(f) and (f’) illustrate spallation zone at higher laser fluence and associated profilometric scan.

Figure 4 (g) illustrates the failure initiation in Al/composite sample. Although no spallation was visible from top, a delamination spot was observed when the layer was manually peeled off after conducting the experiment. At higher laser fluence several cracks in the composite layer appeared (see, Fig. 4(h)). In this sample the carbon ply could not be spalled even after the incidence of the maximum laser fluence.

The interfacial strength of tested specimens is tabulated in Table 2. The data indicates that the adhesion strength of epoxy with aluminum (264 MPa) is higher when compared to the fused quartz substrate (205 MPa). The dynamic strength of FQ/Cu interface is measured to be 146 MPa. The strength of Al/composite adhesion has not been reported since the quantification is still under process.

Table 2. Interfacial failure strength of tested samples

<table>
<thead>
<tr>
<th>Bi-material configuration</th>
<th>( \sigma_{int} ) (MPa)</th>
</tr>
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<tbody>
<tr>
<td>Al/epoxy</td>
<td>263.9(^{+4.1}_{-6.9})</td>
</tr>
<tr>
<td>FQ/epoxy</td>
<td>205(^{+7.2}_{-10.4})</td>
</tr>
<tr>
<td>FQ/Cu</td>
<td>146(^{+9.38}_{-6.43})</td>
</tr>
</tbody>
</table>

6. CONCLUSION

The laser spallation technique is used to characterize the dynamic interface strength of aluminum/epoxy, fused quartz/epoxy, fused quartz/copper, and aluminum/composite specimens. A high amplitude short duration laser induced stress waves are utilized to instigate failure at the bi-material interfaces. Optical micrographs of specimen surfaces are used to identify the minimum laser fluence correspond to the failure initiation. Interfacial failure is confirmed by scanning spallation zone through a profilometer. Optical measurements on calibration samples are performed by employing Michelson interferometry. The substrate stress history assessed from the interferometric data is provided to one dimensional computational wave propagation analysis for inferring thin film interface
strength. The dynamic interface strength of aluminum/epoxy, fused quartz/epoxy and fused quartz/copper is measured to be, 264 MPa, 205 MPa and 146 MPa, respectively.

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