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Strength and Performance Enhancement of Bonded Joints by Spatial Tailoring of Adhesive Compliance via 3D Printing

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Abstract

Adhesive bonding continues to emerge as a preferred route for joining materials with broad applications including advanced structures, microelectronics, biomedical systems, and consumer goods. Here, we study the mechanics of deformation and failure of tensile-loaded single-lap joints with a compliance-tailored adhesive. Tailoring of the adhesive compliance redistributes stresses and strains to reduce both shear and peel concentrations at the ends of the adhesive that determine failure of the joint. Utilizing 3D printing, the modulus of the adhesive is spatially varied along the bondlength. Experimental strength testing, including optical strain mapping, reveals that the strain redistribution results in a greater than 100% increase in strength and toughness concomitant with a 50% increase in strain-to-break, while maintaining joint stiffness. The tailoring demonstrated here is immediately realizable in a broad array of 3D printing applications, and the level of performance...
enhancement suggests that compliance tailoring of the adhesive is a generalizable route for achieving superior performance of joints in other applications, such as advanced structural composites.

Keywords: Adhesive joints, 3D printing, Interface compliance/stiffness tailoring, Graded materials, Polymer composites
1. Introduction

Adhesive bonding is becoming a preferred means of joining materials and structures in the pursuit of realizing cost-effective, lightweight and efficient structural and/or functional systems. However, steep shear and peel stress/strain gradients and concentrations exist at the ends of the overlap causing concern in many applications such as bonded advanced composites, to the point where mechanical fasteners are utilized to supplement the adhesively bonded joint, adding cost, weight, and complexity\textsuperscript{1}. The most commonly occurring configuration to join dissimilar or similar adherends is the single-lap joint (SLJ) due to its ease of manufacture and efficiency of load transfer\textsuperscript{2,3}, although much work has also addressed double lap joints\textsuperscript{4}. In the SLJ, stress/strain shear concentrations in the adhesive layer originate from unequal axial straining of the adherends, and in the case of peel stresses/strains, because of the eccentric load path, concomitantly dictating the failure strength of these joints\textsuperscript{5–7}. Classical stress analysis of SLJ by Volkersen\textsuperscript{8} comprised of two identical elastic adherends of uniform thickness ($t$), and Young’s moduli ($E$) bonded over a length ($l$) by an adhesive of thickness ($h$) with shear modulus ($G$), finds that when bonded members are in pure tension, the shear stress/strain in the adhesive layer is a maximum at each end of the overlap governed by a concentration factor

$$k = \sqrt{\frac{Gl^2}{Eht}} \coth\left(\sqrt{\frac{Gl^2}{Eht}}\right).$$

Volkersen’s analysis suggests that for a given adherend material and geometric configuration, the lower the shear modulus of the adhesive used in the bondline, the lower the stress concentration, leading to higher joint strength. Therefore, in such a configuration (see Fig. 1a), an
adhesive that is very compliant relative to the adherends is commonly utilized to achieve a shear-dominated load transfer across the joint and to minimize the propensity of interfacial and/or cohesive brittle failure due to high peel stresses a relatively stiffer adhesive may experience. While relatively compliant adhesives are attractive, a compromise must be maintained in that joint stiffness has the opposite trend. In many applications, such as advanced structural composites, adhesive resistance to peel stresses is of paramount concern.

Numerous ways of reducing the shear and peel stress concentrations such as modifying the adherend geometry (see, e.g., 10,11), the adhesive geometry (see, e.g., 12) and employing a spew (a bead at the lap ends of the adhesive) geometry (see, e.g., 13,14) have been tried to minimize such stress/strain concentrations to improve the structural response of joints. Research on joints with material-tailored adhesives was pioneered by Hart-Smith 4 and Srinivas 15, towards increasing the structural performance by redistributing the adhesive stresses. Recently, there is growing interest in this area, e.g., it was experimentally shown that the joint strength can be increased about 50-120% by employing a bi-adhesive bondline. 16 The extant work has as of yet considered only a single-step variation in adhesive over the bondlength, with the step-change in both the modulus as well as adhesion energy of the adhesive. However, several recent theoretical studies have considered compliance-tailoring of the adhesive along the bondlength to provide a framework for the mechanics of such bonded systems (see e.g., 17–21). However, manufacturing of bonded systems with spatially varying elastic properties of the
adhesive along the bondlength is challenging, and therefore, the potential of such compliance-tailored adhesive designs have not been explored.

3D printing is a facile approach to realize compliance tailoring of interfaces in multi-material systems so as to produce stronger, tougher and more efficient aerospace/automotive structures, wearable soft-hard prosthetics, energy dissipating structures, gripping mechanisms, robotic arms, multilayer dental and orthopedic implants, among others. Here, we focus on 3D printing of SLJs with compliance-tailored adhesive layers. As shown in Fig. 1a, a SLJ with far-field tensile stress $\sigma_\infty$ contains an eccentric load-path, and both shear and peel stresses/strains are generated in the adhesive, including steep gradients and relatively high absolute values at the ends of the adhesive. The well-known (e.g.,\textsuperscript{9}) typical distribution of adhesive stresses over the bondlength in a SLJ with a constant modulus adhesive is shown in Fig. 1a bottom. Cohesive and/or adhesive failure originates at the stress/strain concentration zones at the adhesive ends leading to catastrophic failure of the joint. Increasing compliance of the adhesive requires a larger length of the bondline to participate in shear stress transfer and has the positive effect of reducing the concentrations at the ends. However, as mentioned earlier, the bondline must be stiff and therefore quick (as measured along the bondlength) load transfer via shear is desired. To this end, we explore two different ways of spatially tailoring the elastic modulus of the adhesive over the bondlength, with moduli profiles shown by the green and orange lines in Fig. 1a center. The compliance tailoring is expected to provide strain tolerance to the joint, reducing stress/strain concentration at the ends, while maintaining load transfer near the center of the joint. The results discussed in Section 3 reveal that SLJs with
compliance-enhanced adhesive ends have a significant increase broadly in performance, including load-bearing capacity.

2. Materials and Methods

2.1 3D Printing

We utilize 3D printing of photopolymers technology that offers the possibility of multi-material deposition\textsuperscript{26,30,31} to print SLJs. Heterogeneous composites that combine stiff reinforcing elements with a soft organic matrix have been successfully exploited for decades in the form of glass or carbon fiber reinforced polymers\textsuperscript{32,33}. Compared to this established technology, multi-material printing offers the possibility of cost-effective automation of the fabrication process and provides greater flexibility to locally design the material architecture in three dimensions\textsuperscript{22,34–44}. The emergence of such multi-material and composites 3D printing technologies facilitates the design of functionally graded designs at sub-millimeter scale enabling fabrication of multi-material structures simultaneously exhibiting both strength and toughness which are often mutually exclusive and thus difficult to reach in homogeneous materials\textsuperscript{45,46}.

CAD models of the joints were created using Solidworks (Dassault Systemes, France) and fabricated using an Object Connex260 Polyjet 3D multi-material printer (Statasys Ltd., USA). Object Connex family 3D printers can print two different polymers simultaneously and could combine these two basic polymers in different proportions to produce a range of materials called digital materials. This printer deposits the polymeric material via an array of 8 print heads that sprays the liquid polymer, subsequently the polymer is cured by UV light,
before the next layer is added. The nozzles are mounted on the print head which moves in the \( x \) and \( y \) directions. The print head only jets the liquid polymer when it is moving across the tray in the \( x \) direction. Therefore, the printing direction could influence the material behavior of printed specimens\(^{47}\), although this is avoided in the comparisons undertaken here. A support material (Objet Support SUP705) was used during printing to enable fabrication of overhang portions of geometries and was subsequently removed through water washing. The resolution of the 3D multi-material printer is 16 \( \mu \)m in the \( z \) direction (thickness) and 42 \( \mu \)m in the \( x \) and \( y \) directions. Therefore, the smallest geometric feature of the joints was designed to be at least an order of magnitude greater in size than the resolution of the 3D printer to minimize any effects of the 3D printing process on the mechanical response\(^{48}\).

2.2 Adhesive Tailoring

The compliance tailoring approach adopted here takes advantage of the 3D printing capabilities to control the relative volume fraction of polymer\(^{26,30,49}\) so as to modify the compliance of the adhesive while maintaining the same material interfaces between the adherends and the adhesive. The adherends were 3D printed using a rigid polymer VeroWhitePlus\(^{\text{TM}}\) RGD835 (VW) and it's Young’s modulus (\( E_a \)) is 2250 MPa. The adhesive was printed using both TangoPlus\(^{\text{TM}}\) FLX930 (TP), and a digital material S40 having Young’s moduli \( E_1 = 0.536 \) MPa and \( E_2 = 1.098 \) MPa, respectively\(^{47,48}\). See discussion and Fig. S1 for properties of the different polymers, principally noting that \( E_a >> E_1 \approx 0.5E_2 \). The different configurations tested are shown in Fig. 1b, and include both stiffness (in the center) and compliance (at the ends) addition.
to the base adhesive by printing widthwise-constant circular features into the adhesive in a staggered pattern. Preliminary testing looking at different distributions and shapes of features (see Figs. S3 and S4 and associated discussion) to impart compliance change were investigated leading to the geometries and materials used herein. A refined version of preliminary Design 1 shown in Fig. S3a with $l_2 = 22.14 \text{mm}$, $L = 50 \text{mm}$ and number of circular features $n_i = 20$ were considered for center stiffness tailored adhesive (see Fig. S5a) with local volume fraction of circular features in the central region of 15\%, giving a local modulus change of +16\% versus the $E_1$ adhesive. Similarly, an improved version of preliminary designs shown in Fig. S4a with $l_1 = l_3 = 9.53 \text{mm}$, $L = 50 \text{mm}$ and number of soft circular features $n_i = 7$ at each end zone were considered for the edge compliance tailored adhesive (see Fig S5b) with local volume fraction of circular features in the end zone being 15\%, giving a local modulus change of -7\% versus the stiffer $E_2$ adhesive. In addition, adhesives that have these features along the entire bondlength for both cases were considered. The 3D printed SLJs were tested on a Zwick-Roell tensile testing machine with a 2.5kN load cell. The load was applied at a constant speed of 5 mm/minute. Digital image correlation (DIC) was used to map the two-dimensional state of deformation and calculate strain in the joint as a function of load. Random speckle patterns were formed on the specimen’s surface prior to testing by applying a thin coat of white acrylic paint followed by spraying random dots of black acrylic paint using an airbrush. A monochrome 5.0 MP camera was used to capture the speckle images during the test. Vic-2D
software was used to compute the evolution of the strain field as a function of load. Each of the SLJ designs was tested three times to ensure repeatability.

**Figure 1.** Spatially-tailored adhesive compliance in single-lap joint (SLJ): (a) Conceptual representation of SLJ subjected to a far-field tensile stress $\sigma_\infty$ with different adhesive modulus profiles. Bottom shows stress distributions in the adhesive for a constant modulus adhesive (either $E_1$ or $E_2$). (b) Optical images of 3D printed joints with adhesive compliance tailoring considering all four profiles in a), including center stiffness- and edge compliance-tailored cases. The thickness of the adhesive in all cases is 7mm. Note that $E_1 \approx 0.5E_2$.

**Figure 2.** Performance of tailored- and constant-modulus adhesive joints: (a) Load-displacement curves using compliant adhesive with modulus $E_1$, and (b) with modulus $E_2$. Note that $E_1 \approx 0.5E_2$. 
3. Results and Discussion

**Table 1.** Summary performance of 3D printed tailored and non-tailored joints.

Changes (shown in red) are calculated vs. the constant modulus adhesive in each case, and only statistically significant increases are shown.

<table>
<thead>
<tr>
<th>Design Configuration</th>
<th>Joint stiffness (N/mm)</th>
<th>Maximum load (N)</th>
<th>Deflection at break (mm)</th>
<th>Toughness ($\times 10^3$ N*mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_1$ adhesive</td>
<td>26.8 ± 0.493</td>
<td>231 ± 9.39</td>
<td>9.70 ± 0.163</td>
<td>1.24 ± 0.07</td>
</tr>
<tr>
<td>$E_1$ tailored with center stiffness</td>
<td>29.1 ± 0.295 (8.58%)</td>
<td>443 ± 7.87 (91.8%)</td>
<td>14.6 ± 0.142 (50.5%)</td>
<td>3.34 ± 0.076 (169%)</td>
</tr>
<tr>
<td>$E_1$ non-tailored (stiffness added)</td>
<td>31.6 ± 0.03 (17.9%)</td>
<td>263 ± 6.59 (13.9%)</td>
<td>9.33 ± 0.226 (9.68%)</td>
<td>1.36 ± 0.067 (9.68%)</td>
</tr>
<tr>
<td>$E_2$ adhesive ($E_2$~2*$E_1$)</td>
<td>44.9 ± 0.375</td>
<td>403 ± 3.33</td>
<td>9.21 ± 0.07</td>
<td>1.95 ± 0.03</td>
</tr>
<tr>
<td>$E_2$ tailored with edge compliance</td>
<td>47.2 ± 0.316 (5.12%)</td>
<td>848 ± 24.3 (110%)</td>
<td>13.7 ± 0.281 (48.8%)</td>
<td>5.24 ± 0.213 (169%)</td>
</tr>
<tr>
<td>$E_2$ non-tailored (compliance added)</td>
<td>45.4 ± 0.19 (70.2%)</td>
<td>686 ± 10.9 (37.9%)</td>
<td>12.7 ± 0.130 (110%)</td>
<td>4.10 ± 0.098 (110%)</td>
</tr>
</tbody>
</table>

Representative load-displacement responses are shown in Fig. 2 for the four SLJ configurations in Fig. 1b. As expected, the relatively stiffer and stronger adhesive imparts a stiffer and also stronger (~2×) SLJ response than the more compliant adhesive, given the geometric parameters considered. It can also be seen that both tailoring approaches have merit relative to the constant-modulus adhesives, where joint stiffness is maintained and strength is enhanced (see summary data in Table 1). The compliance tailored ends of the adhesive in Fig. 2b is shown to be the highest performing across all relevant metrics, achieving a large strength (110% increase relative to the homogeneous adhesive $E_2$ and even greater compared to homogeneous adhesive $E_1$), and toughness increase while maintaining joint stiffness (see Table 1). Importantly, all
configurations tested contain the same adhesive-adherend interfaces – unlike prior work that varies both the modulus and the energy of adhesion of the adhesive and the adherend by tailoring the adhesive in a step-wise fashion. Thus, in the work herein, we can conclude that the increased performance is (solely) attributable to stress/strain redistribution due to spatially tailoring compliance. This is explored further in strain imaging for the specimens tested.

It should be noted that the case of non-spatially tailored features (where stiffness or compliance is added over the entire bondlength) is also presented in Table 1, with discussion to follow.

The improved joint performance in Fig. 2 and Table 1 is due to the redistribution of stress and strain in the adhesive as a direct result of the compliance tailoring along the bondlength. At loads where the response remains linear elastic, the strain re-distribution is evident in Figs. 3 and 4 for the various modulus profiles. Note that in Fig. 3, the $E_2$ circular features add stiffness to the $E_1$ adhesive, whereas in Fig. 4, the $E_1$ features add compliance to the $E_2$ adhesive. In Fig. 3, the peel and shear strain distributions are shown at a load of 50 N (see Fig. 2a) and the trend is consistent with expectations: (1) the compliant adhesive (relative to the adherend) in the top row transfers the load in shear with the development of strain concentrations at the free edges of the adhesive; (2) adding stiffness across the entire bondlength (see bottom row of Fig. 3 where $E_2$ features are added) maintains the same distributions but reduces the magnitude. This is also similar to the $E_2$ adhesive case in the top row of Fig. 4, as expected although the magnitudes are again reduced. The
distributions in Figs. 3 and 4 are noted to agree with the expected behavior of compliant adhesive SLJs.

Even at the relatively low load and strain levels in Figs. 3 & 4, the effect of the features (center stiffness added, and edge compliance added, respectively) are noted to redistribute strains in the adhesive layer, and reduce magnitudes of both at the critical end zones of the adhesive. The compliance tailoring acts to reduce strains overall, both shear and peel, particularly in the critical high-strain regions at the adhesive ends (compare the top and middle rows in Fig. 3), with similar behavior in Fig. 4. The influence of spatial tailoring is revealed in higher values of loading and strain as shown in the strain maps in Figs. 5 (at 130 N load) and 6 (at 237 N load) and the failure images in Figs. 7 & 8, for the center stiffness and edge compliance- tailored cases, respectively. Here, the redistribution of strains can be seen in both tailored cases (center rows in Figs. 5 & 6), in particular reducing strain concentrations (both shear and peel) near the adhesive edges. It is worth noting that the strain maps for the (unfailed) compliance tailored adhesives shown in Figs 5 and 6 (middle rows) correspond to a load level at which failure initiates in constant moduli adhesives (top rows of Figs 5 and 6).

Joint failure (focusing on loss of load-carrying capability, as in ultimate strength) can be observed in the strain map shown in Fig. 6 for the constant modulus $E_2$ joint where a crack forms near the adhesive-adherend interface. Indeed, in all cases failure is observed as the initiation and growth of a crack in the region of strain/stress concentration at the adhesive ends. From Figs 7, 8, S7 and videos of their failure (see Supporting Information), we observe that in all cases
macroscopic failure near the peak load emanates as a crack close to the corner either at the adhesive-adherend interface (adhesive failure) or within the adhesive (cohesive failure) very close to the bondline. The homogeneous adhesive $E_1$ and $E_2$ cases are exemplary, in that the emanated failure propagates very quickly as adhesive failure over the entire bondlength. The purpose of our study is not to consider or engineer tougher behavior, but rather to highlight the effects of spatial compliance tailoring of the adhesive to reduce critical strains (shear and peel) driving failure at the adhesive ends. Noting that toughness is influenced by the relative strengths and toughness of the $E_1$ and $E_2$ materials, as well as by compositing effects of the features and new failure modes such as feature debonding (all of which are uncontrolled in the current study), we instead focus on strength with toughness reported in Table 1 simply for completeness. In all cases, the peak load is associated with a crack appearing and propagating at the peak load, such that comparing relative strengths of the joints due to compliance tailoring can be made.

Figure 3. Strain field distribution in 3D printed SLJ with/without stiff center features under tensile load by DIC at 50 N (see Fig 2a): a), schematic of the adhesive, b) shear strain, and c) peel strain in the joint, respectively. $E_1$ is the modulus of the adhesive and $E_2$ is the modulus of the stiffer circular features.
Figure 4. Strain field distribution in 3D printed SLJ with/without compliant edge features under tensile load by DIC at 200 N (see Fig 2b): a), schematic of the adhesive, b) shear strain, and c) peel strain in the joint, respectively. $E_2$ is the modulus of the adhesive and $E_1$ is the modulus of the soft circular features.

Figure 5. Strain field distribution in 3D printed SLJ with/without center stiffness features under tensile load by DIC at 130 N (see Fig 2a): a), schematic of the adhesive, b) shear strain, and c) peel strain in the joint, respectively. $E_1$ is the modulus of the adhesive and $E_2$ is the modulus of the stiffer circular features.
Figure 6. Strain field distribution in 3D printed SLJ with/without edge compliance features under tensile load by DIC at 247 N (see Fig 2b): a), schematic of the adhesive, b) shear strain, and c) peel strain in the joint, respectively. $E_2$ is the modulus of the adhesive and $E_1$ is the modulus of the soft circular features.

Figure 7. Deformations near failure in 3D printed SLJ with/without center stiffness features under tensile load: a), representative load-deflection response, and specimens under loading at b) point A, c) point B, and d) point C in Fig. 7a.

Figure 8. Deformations near failure in 3D printed SLJ with/without edge compliance features under tensile load: a), representative load-deflection response, and specimens under loading at b) point A, c) point B, and d) point C in Fig. 8a.
4. Conclusions

In this study, compliance-tailored adhesive designs are explored via 3D printing to illustrate the potential of such an approach towards generating superior multi-material adhesive interfaces. The dependence of (single-lap) joint performance, focusing on strength while maintaining stiffness, was found through experiments including optical strain mapping. While retaining joint stiffness, more than a 100% increase in strength and 150% increase in toughness were observed for both center stiffness and edge compliance tailored adhesive designs of the joints in comparison with their non-tailored (stiffness/compliance added) and constant moduli counterparts due to reduction in peak peel and shear strains at the ends of the adhesive. The approach of compliance-tailoring demonstrated here is quite general and immediately realizable and useful in both current (e.g., multi-polymer 3D printers as used here) and emerging (polymer, metal, ceramic) multi-material 3D printers, e.g.,28,35,50–54. In addition to joining in multi-material additive manufacturing, the approach may be applied to joining separately additively manufactured structures, and other joining problems such as advanced composites. If the correct levels of spatial tailoring in the adhesives can be achieved in structural composite joining, perhaps performance can approach what some have called optimal design of such interfaces by bio-mimicking55–61. Although not explored in the current study, tailoring of the adhesive can also impart new mechanisms of toughness to joints that allow some load-carrying capability beyond the critical load, by imparting the usual composite toughening mechanisms, such as localized shear banding leading to increase in tortuosity of the crack path11,62–70. The change in failure mode/mechanism and delay in
growth of multi-site cracks due to tailoring may provide design opportunities to increase both strength and toughness, properties that are often mutually exclusive.

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Supporting Information

Supporting information includes material and geometric properties of the 3D printed joints, preliminary results for different cases of compliance tailored adhesive joints, fracture images, and synchronized videos of load-displacement curves and deformation maps of representative 3D printed SLJs (SV1 and SV2).

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