EFFECT OF SURFACE CONTAMINATION WITH MITIGATION METHODS ON ADHESIVE COMPOSITE BOND INTEGRITY AND DURABILITY

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ABSTRACT

Adhesive bonds offer multiple advantages over traditional fasteners such as reduced weight and a potential decrease of stress concentrations due to hole fabrication. However, the influence of undesirable bonding conditions on the strength of adhesive bonds such as bonding surface contamination has not been reliably characterized. In this study, a method was proposed to create a scalable weak bond using a continuous spray approach. In this approach, the mass of a silicon based mold release can be scaled to achieve various levels of contaminant effect. Methods to mitigate the effect of contamination such as sanding and solvent wiping were investigated, and repeated using environmental exposure to investigate bond durability. To assess bond strength, Dual Cantilever Beam (DCB) tests were performed and the critical energy release rate was measured. Results showed the critical energy release rate of the bonded joints correlated well with the mass of the contaminant applied to the surface. Results of the mitigation procedures showed that solvent wipes led to minimal statistical changes. However, a combination of wipe-sand-wipe procedure yielded a partially recovered load capacity and failure mode. Environmental exposure testing revealed no added combinatory effect with the addition of the contaminate on the surface.

1. INTRODUCTION

1.1 Background

There have been ever increasing demands in the aerospace industry for bonding technologies where the weight savings is a key driving force for an aircraft. Currently, aircraft primary structures are mostly composed of metals such as aluminum, steel, or titanium in order to support heavy loads. While the goal is for the craft to be as light as possible, it must also still be able to support the same load and perform at the same level as the current standard. The need for high strength to weight ratio creates a demand for composite materials in the aerospace industry due to their exceptional specific strength and stiffness (on the order of 100 MNm/kg for stiffness and 500 kN/kg for strength). However, joining composite materials with conventional mechanical fasteners just as metals are causes delamination in the composite panels (such as in Figure 1) and creates stress concentrations. These stress concentrations often ultimately become the point of origin for failures under load. The proposed solution is to use adhesively bonded composite joints to replace mechanically fastened metals on primary structures, thus achieving the goal of reducing weight

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without sacrificing strength of the material. Unfortunately, there are some issues with adhesive bonding. During the manufacturing and handling of the panels, the surface can become contaminated, which can significantly reduce the integrity of the bond. The effects of harsh environmental conditions on the bond have not been quantified, and thus it is difficult to predict their effects. Finally, bonded composites tend to fail in a brittle nature and it is very complicated to nondestructively predict this failure.

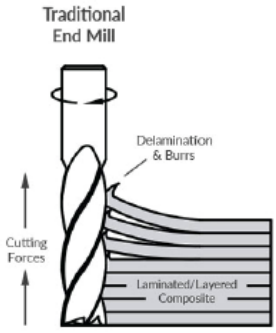


Figure 1: Delamination in composite panel due to mechanical fasteners.[1]

1.2 Motivation

Past research has revealed that contamination at the bond interface significantly influences bond quality and mechanical response of adhesively bonded joints. Contamination could result at various stages of manufacturing, including during the application and removal of peel ply material for surface preparation of the composite surface[2,3,4] and from the easy release ply sheet from the adhesive tape[5]. There exists a need to quantify the levels of contamination that affect the bond strength, as well as the magnitude of the effect on bond strength, in order to determine the acceptable limits of contamination that a bonded joint can sustain without premature failure. In the efforts of quantification, Van Voast et. al[6] introduced contaminate at on the peel ply at various levels to determine their effect on bond strength and concluded that a 2% siloxane level of contamination could significantly affect adhesion. The release properties of various release agents/coatings were studied and an effective reduction in bond strength with Frekote contaminant was observed by Critchlow et. al[7]. Composite panels were dipped into Frekote (4 at % Si) showed a 70% reduction in fracture toughness[8]. Jeenjitkaew et. al[9] contaminated the interface between a metal and adhesive bond on a double lap joint and observed a 27% reduction in the bond strength. Although the link between contaminant and bond quality has been established, there is a lack of repeatability of methods to evaluate the effect of contamination on an adhesive joint. In addition, the initiation location and progression of damage in contaminated regions remains a mystery.

1.3 Goals

Ultimately, the goal of this research is to investigate the effect of surface contamination on the integrity of adhesively bonded composite joints and what – if any – effect do mitigation techniques have on restoring bond strength after contamination. These weak bonds need to be scalable in order to be able to test the extent of the contamination effect at different amounts of contaminant applied. Using this approach, mitigation techniques can also be evaluated for their ability to return the bond to its nominal state. Finally, the effect of aging factors such as environmental exposure are investigated and quantified.

2. EXPERIMENTATION

2.1 Specimen Fabrication

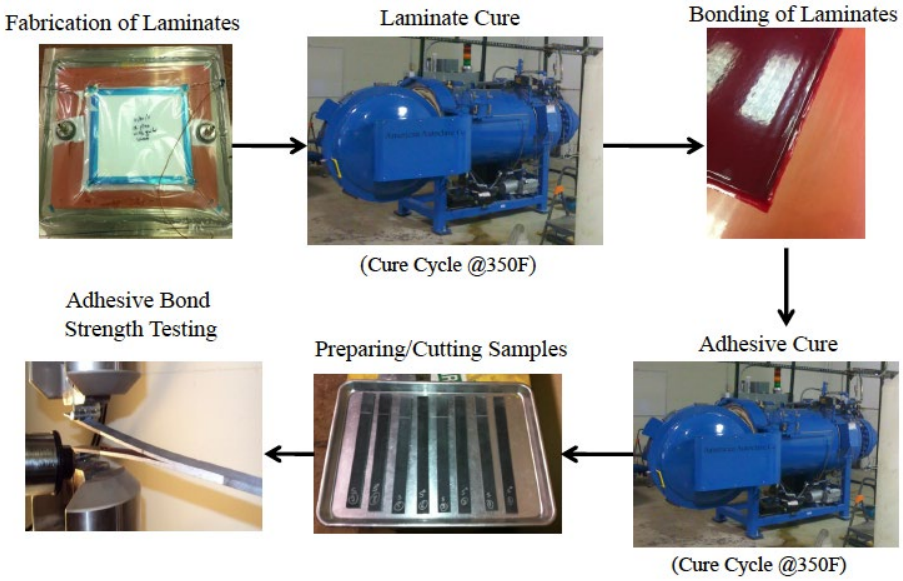


Figure 2: Adhesively bonded composite joint manufacturing process.

Dual Cantilever Beam (DCB) test coupons were manufactured to adhere to ASTM D5528 standard with the feasibility of testing specimen dimensions. The materials list included:

- Prepreg- T800 Unidirectional carbon fiber (P2362W-19U-304) from Toray
- Adhesive- AF 555M film adhesive from 3M
- Peel ply- 60001 polyester ply from Precision Fabric

The manufacturing process in its entirety can be seen in Figure 2. Laminates each consisted of 10 plies with 0º ply orientation and were cured in an autoclave at 177 ºC (350 ºF) maintaining a vessel pressure of 207 kPa (30 psi). The cured laminates were bonded using a secondary bonding procedure where two ply adhesive films were utilized. Post bonding, macro-scale coupons for dual cantilever beam (DCB) testing were prepared with dimensions 254 mm (10 in) in length and 25.4 mm (1 in) in width with a pre-crack of 63.5 mm (2.5 in). Bondline thickness measurements were obtained using an optical microscope. Measurements were taken every centimeter for the first 5 cm starting from the pre-crack of the DCB coupons. Ten readings per coupon were measured and the bondline reported is the average of these measurements for each specimen.

2.2 Contamination Procedure

The contaminate used in the study (Frekote 700-NC, Henkel Corporation) is based on Poly Di-Methyl Siloxane (PDMS), was chosen as a result of its prevalence of use in traditional composite manufacturing environments. Frekote works by inhibiting the hydrogen bonding that typically occurs during amine adhesive curing. Previous studies using Frekote 700-NC have shown the ability to locally limit adhesive bonding when applied resulting in weak bonds at the adhesive interface.[8]

For the uniform contamination approach, a nozzle mounted on a moving head was used to uniformly spray contaminant across the surface of the laminate. This was done by building a computer-controlled machine consisting of a 2-axis stepper motor controller Arduino that uses computer-generated code to move the nozzle in repeated and consistent patterns along the XY plane. The complete setup is shown in Figure 3. In this case, the nozzle was programmed to follow a linear spray pattern of 8 inches along the x-axis and ½ an inch along the y-axis repeatedly throughout the longitudinal direction of the laminate. The width of the spray pattern was adjusted for even coverage of the panel with minimal overspray between each horizontal run. Gravimetric analysis was performed before and after contamination in order to monitor the amount of contaminant sprayed on the laminates. The uniform patterned horizontal bands of sprayed contaminant on a composite surface can be viewed in Figure 4.

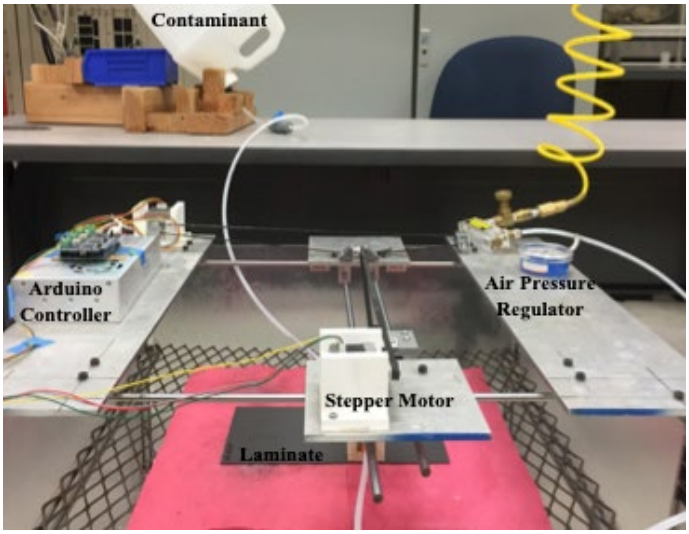


Figure 3: "Contamination station" used to distribute contaminant evenly in a scalable fashion.

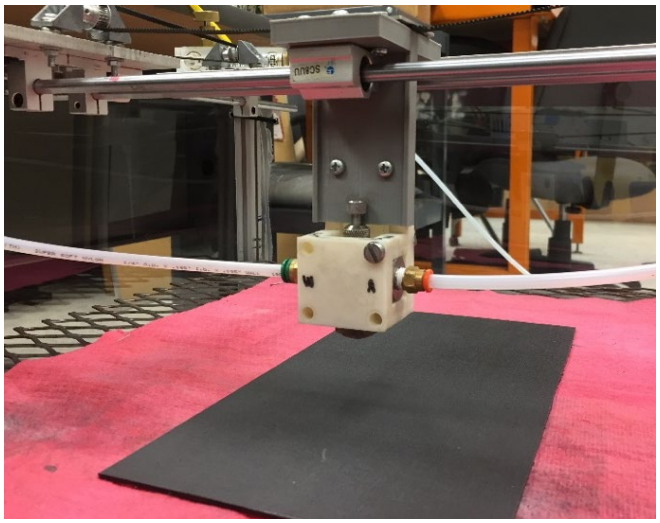


Figure 4: Close-up of apparatus used to contaminate composite panels.

Calibration of the contamination levels was completed in order to be able to trace back the amount of contaminant used and relate that amount to the strength of the weak bond created. In this case,

one level of contamination was evaluated which would correlate to a medium level bond strength between 40-60% reduction in the baseline. These percentages represent the bond strength recovery compared to the bond strength of the controlled non-contaminated samples. The amount of contaminant sprayed onto the laminate was controlled by adjusting the spray speeds and size of the nozzle with a fixed air pressure of 15 psi and a fixed height of 1 inch from the surface of the composite to the nozzle head. Measurements were done of the mass of the contaminant on a 1" x 1" section of a panel before and after every level of contamination to further create a relationship between the mass of contaminant on the composite surface and the bond strength created from that mass. This procedure was done for calibration purposes for several contamination levels in order to be able to classify the different bond strengths that can be created from the different amounts of contaminant.

2.3 Mitigation Methods

Once the laminates have been contaminated, the focus was shifted to mitigating the effects. Two methods selected were a solvent wipe technique and a wipe/sand/wipe technique, as shown in Figure 5. Both methods are typical mitigation procedures found in small OEM facilities. The solvent wipe technique is simply using a solvent wipe (SATWIPES Wipes Presaturated with 100% MPK) to attempt to remove the contaminant from the surface of the laminate. The wipe/sand/wipe technique attempts to remove the contaminant from the surface of the laminate using a solvent wipe, then using an orbital sander, followed by one more round of solvent wipe. The orbital sander is a Dynabrade 2" Dia. Right Angle Disc Sander, using a 180 Grit Aluminum Oxide Quick Change Discs Type 3 abrasive from Merit Abrasives.

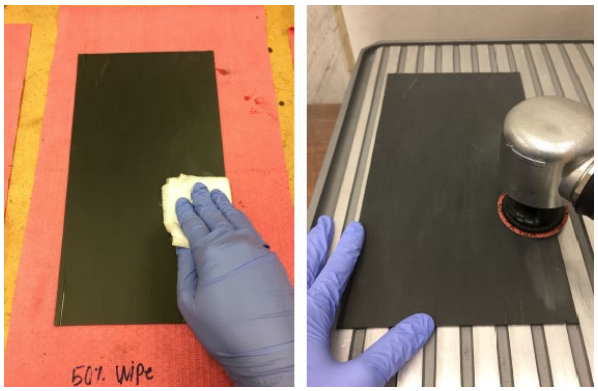


Figure 5: Mitigation procedures selected: Wipe (left) and wipe/sand/wipe (right).

2.4 Environmental Exposure

In addition to contamination, some coupons were selected to be exposed to hot and wet conditions in an environmental chamber for 8 weeks. 8 coupons were manufactured for each set: baseline, contaminated, and wipe/sand/wipe. 4 coupons from each set were exposed in the environmental chamber and the remaining 4 coupons served as the unexposed set. After 8 weeks in the environmental chamber set to 50°C and 95% relative humidity, the exposed samples were removed from the chamber and DCB tests were performed.

2.5 Testing

The results of DCB tests were conducted as per ASTM D 5528. DCB testing was conducted using an MTS Criterion 43 manufactured by MTS. The loading rate was applied at a rate of 5 mm/min. With the increments in the loading rate, the delamination length (crack length) was measured on

one side of test specimen with the aid of an optical microscope camera. The delamination onset was marked with the help of a silver marker starting from the end of crack insert and gradations were made for every 1 mm for the first 5 mm and continuing the vertical gradation for every 5 mm from thereon (ASTM D 5528).

In addition to quantitative analysis, qualitative observations of failure modes of each group of samples was conducted. Failure modes can be indicators of what effect the contaminant is having on the adhesive joint and how effective the mitigation techniques are at recovering lost bond strength. Different failure modes are shown in Figure 6.

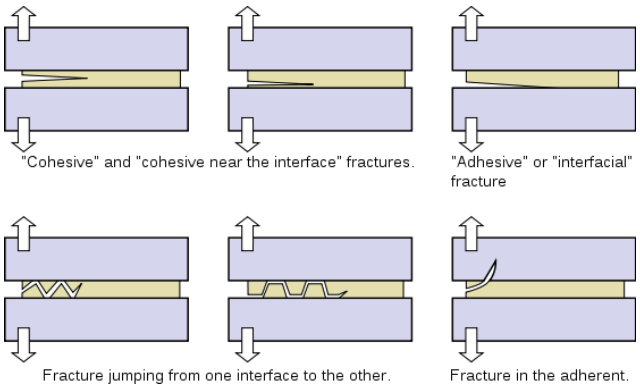


Figure 6: Different failure modes for adhesively bonded composite joints.[10]

3. RESULTS

3.1 Contamination and Mitigation Results

3.1.1 Fracture Properties

The critical energy release rate of the bonded coupons for Mode I propagation were investigated using DCB Testing. These were conducted for baseline coupons, as well as contaminated coupons with no mitigation methods, the wipe method, and the wipe/sand/wipe method. The baseline specimens (whose G_{1C}s are shown in Figure 7) exhibited an average G_{1C} value of 0.87 kJ/m^2, while the contaminated set (Figure 8) exhibited an average G_{1C} value of 0.39 kJ/m^2 – about 42% of the bond strength of the baseline coupons.

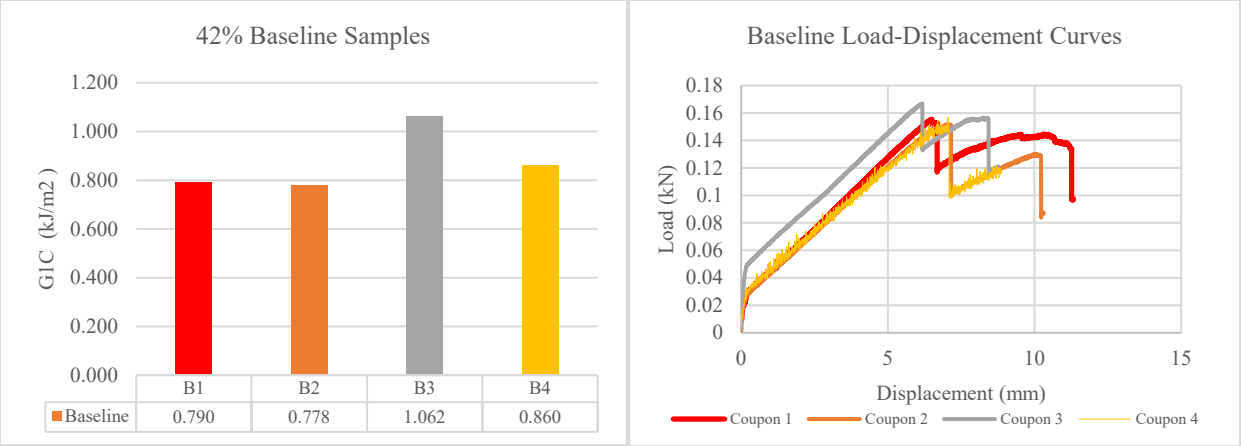


Figure 7: DCB test results for baseline coupons.

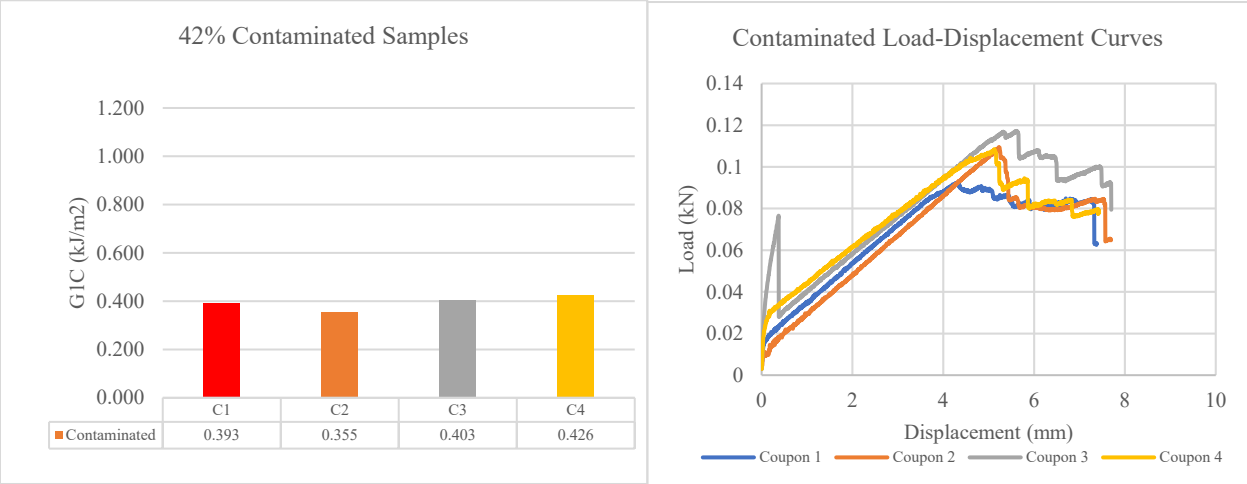


Figure 8: DCB test results for contaminated coupons.

Results of the mitigation procedures are shown in Figure 9 and Figure 10. The wiped specimens exhibited an average G_{1C} of 0.40 kJ/m^2, which is about 43% of the bond strength of the non-contaminated baseline specimens (only about a 1% increase from the unmitigated contaminated specimens). The wipe/sand/wiped specimens showed an average G_{1C} of 0.82 kJ/m^2, which is about 96% of the bond strength of the baseline samples. This method was able to recover approximately 54% of the bond strength lost from the weak bonds created by the Frekote contaminant.

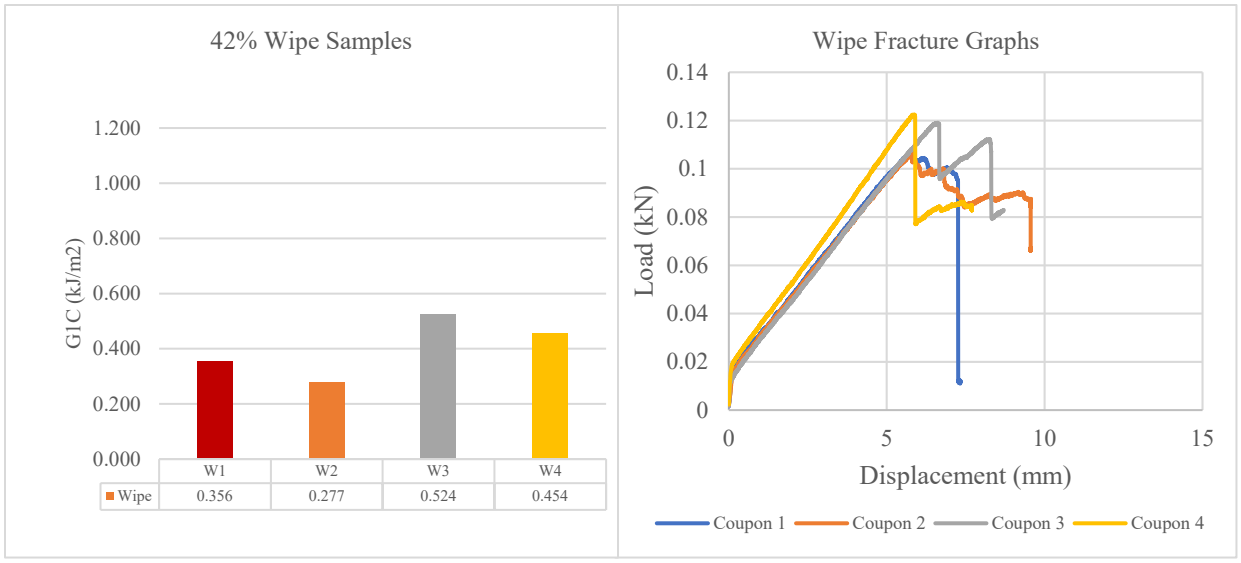


Figure 9: DCB test results for wipe coupons.

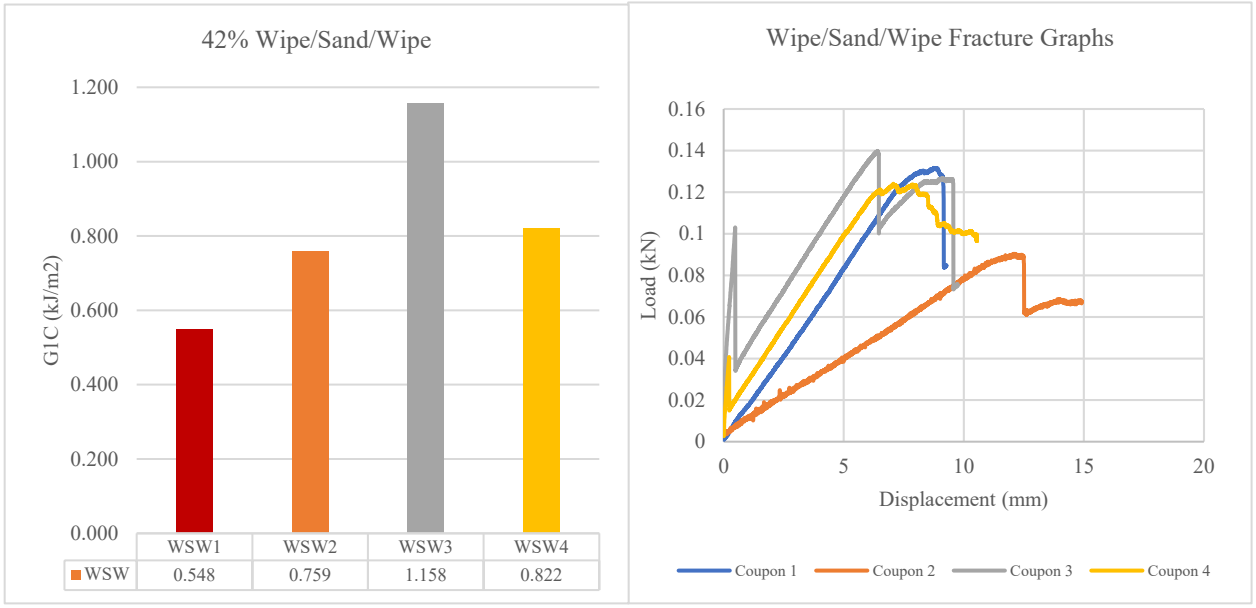


Figure 10: DCB test results for wipe/sand/wipe coupons.

As can be seen in the summary of results (Figure 11), contamination had a tremendous effect on the integrity of the adhesive bonding. While the wipe/sand/wipe method was very successful in recovering lost bond strength due to contamination, the wipe technique was unable to recover any of the original bond integrity. The mitigation procedures also produced a bit more variation in the resulting G_{1C} values, which is most likely due slight differneces in the sanding and wiping application.

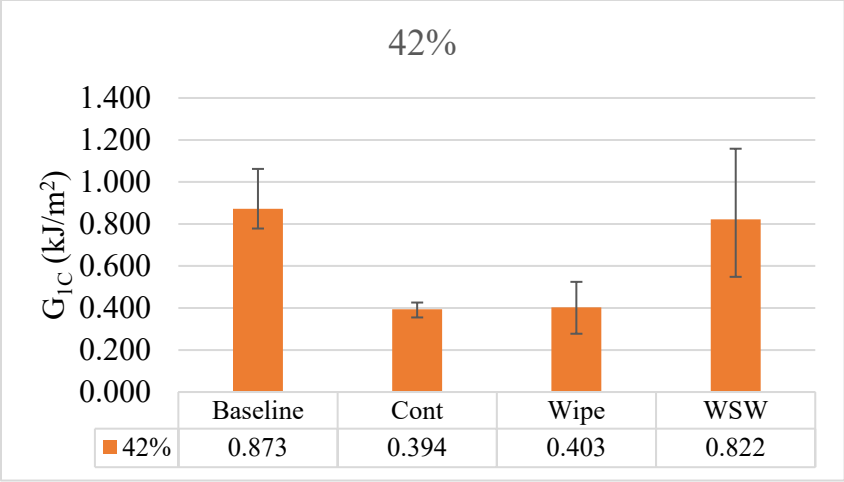


Figure 11: Summary of contamination and mitigation results.

3.1.2 Failure Modes

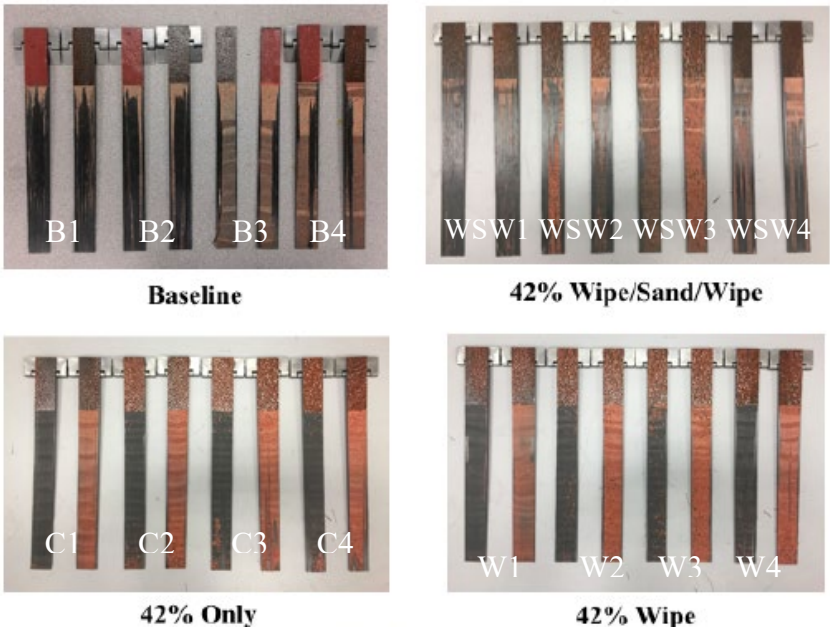


Figure 12: Failure modes of all four sets of coupons.

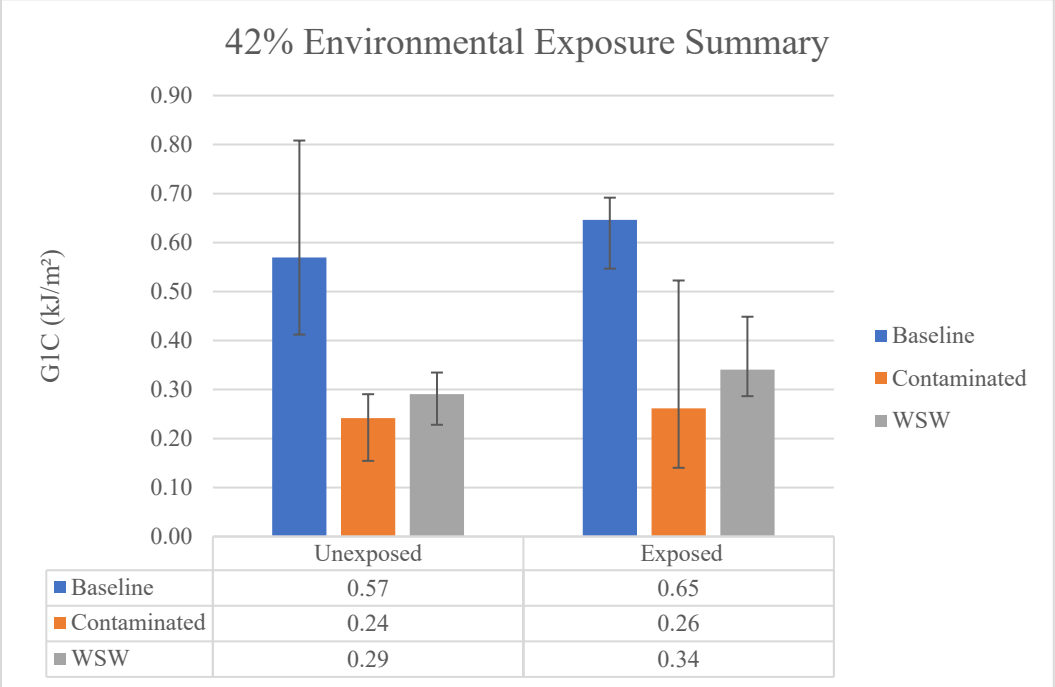
Figure 12 shows the failure modes of all four sets of DCB samples. The unmitigated contaminated samples exhibited properties of pure adhesion failure, meaning that the adhesive separated from the surface of the adherent. This implies that the contamination was successful in creating weak enough bonds such that the adhesive failed at the interface as expected. The wipe technique sample set also exhibited adhesive failure, which supports the results of the DCB testing yielding a similar G_{1C} to the unmitigated samples. Both the baseline and wipe/sand/wipe sample sets display mixed-mode failure - a combination between interlaminar and cohesive failure – which is verified by the similar fracture toughness values. This type of failure requires more energy to take place, which again is backed up by the stark contrast in G_{1C} values between the baseline and wipe/sand/wipe sets vs the unmitigated and wipe technique sets. For example, looking at the B3 coupon (third pair

from the left) it can be seen that there is adhesive on both halves of the sample, implying that a cohesive failure took place (Figure 6). Since B3 is the only coupon of the set that has a pure cohesive failure, it should have the highest fracture toughness - which is confirmed by Figure 7.

3.2 Environmental Durability Results

DCB testing was used in the investigation of environmental durability as well. These tests were conducted for baseline coupons, as well as contaminated coupons with no mitigation methods, and the wipe/sand/wipe method, and were carried out with the same parameters as the tests in 3.1.1. The summary of results is shown in Figure 13.

For the unexposed sample set, the baseline specimens exhibited an average G_{1C} value of 0.57 kJ/m^2, while the contaminated set exhibited an average G_{1C} value of 0.24 kJ/m^2 – about 42% of the bond strength of the baseline coupons. Finally, the wipe/sand/wipe technique sample set showed an average G_{1C} value of 0.29 kJ/m^2 (50% of baseline bond strength). In the exposed sample set, the baseline specimens yielded an average G_{1C} of 0.65 kJ/m^2, which is slightly stronger than their unexposed counterparts. Contaminated samples exhibiting 0.26 kJ/m^2 (about 46% of unexposed baseline bond strength), and wipe/sand/wipe samples at 0.34 kJ/m^2 (about 60% of unexposed baseline bond strength). The wipe/sand/wipe technique appeared to recover some of the strength lost to contamination, but not nearly as significant as in the previous study.



	Unexposed	Exposed
Baseline	0.57	0.65
Contaminated	0.24	0.26
WSW	0.29	0.34

Figure 13: Summary of environmental exposure study results.

4. CONCLUSIONS

In this study, the influence of surface contamination on bond quality was investigated through the development of a scalable surface contamination procedure. As the demand for composites in the aerospace industry continues to grow it is increasingly important to understand the additional factors that may influence bond performance. In addition, the development of an effective procedure to mitigate the effect of the contaminant on bond quality becomes imperative. It was evident that mitigation procedures can be a successful tool to assure that contaminated panels do not go to waste. The failure mode analysis helped in understanding the role of contamination in governing the exactly how bonded joints yield. Finally, the environmental exposure study showed that over an 8-week period, bond strength does not yield by a significant amount. In fact, G_{1C} values seemed to increase across the board.

Ultimately, the results of this study can be used to improve the design of adhesively bonded composites and to design NDE techniques to assess bond quality prior to implementation. For instance, hand-held FTIR scans can be used at various stages of composite processing/composite bonding to determine the presence of contaminate locations and overall contaminated area. Standards for acceptable amount of contamination can focus solely on the mass of the total contaminant applied. Mitigation procedures that involve wipe/sand/wipe or similar techniques may be useful in recovering lost bond strength from the contamination. In addition, future design can incorporate direct evidence of the influence of weak bonds on overall performance in design and modeling software, leading to enhanced confidence in predictions and a reduction overall component weight and in the need for arbitrarily high safety factors or overdesigned mechanical fasteners.

5. REFERENCES

1. Harvey Tool. Harvey Tool - Carbide Composite Cutting End Mills - Compression Cutter. N.D. 19 2 2019. Http://Www.Harveytool.Com/Secure/Content/Images/Compressioncutter.Jpg>.

2. L. Hart-Smith, "Adhesive Bonding of Aircraft Primary Structures," in *Aerospace Congress & Exposition*, Long Beach, Calif, 1980.

3. L. Hart-Smith, G. Redmond and M. Davis, "The Curse of Nylon Peel PLy," in *41st International SAMPE Symposium*, 1996.

4. D. Klapprott, H. Li, R. Wong and G. Geisendorfer, "Key Factors of the Peel Ply Surface Preparation Process," Bay Point, California, USA, March 2004.

5. J. Scheirs, Modern Fluoropolymers: High Performance Polymers for Diverse Applications, Victoria, Australia: John Wiley & Sons Ltd, February 2000.

6. P. Van Voast, P. Shelley, P. Blakley, A. Tracey, B. Flinn, G. Dillingham and B. Oakley, "Effect of Varying Levels of Peel Ply Contamination on Adhesion Threshold," in *Society for the Advancement of Material Process and Engineering (SAMPE)*, Seattle, WA, 2010.

7. G. Critchlow, R. Litchfield, I. Sutherland, D. Grandy and S. Wilson, "A review and comparative study of release coatings for optimised abhesion in resin transfer moulding applications," *International Journal of Adhesion & Adhesives,* vol. 26, pp. 577-599, 2006.

8. D. Markatos, K. Tserpes, E. Rau, S. Markus, B. Ehrhart and S. Pantelakis, "The effects of manufacturing-induced and in-service related bonding quality reduction on the mode-I fracture toughness of composite bonded joints for aeronautical use," *Composite: Part B,* vol. 45, pp. 556-564, 2013.

9. C. Jeenjitkaew, Z. Luklinska and F. Guild, "Morphology and surface chemistry of kissing bonds in adhesive joints produced by surface contamination," *International Journal of Adhesion and Adhesives,* vol. 30, pp. 643-653, June 2010.

10. Brandtner-Hafner, Martin. "Interface Fracture Mechanics of Notched Wood-Adhesive Composites at Mode I, II, III Loading." *International Symposium on Notch Fracture*. Santander: ResearchGate, 2017.