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# Effect of Surface Contamination on Composite Bond Integrity and Durability

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# Composite Bond Integrity/Long-Term Durability of Composite Bonds

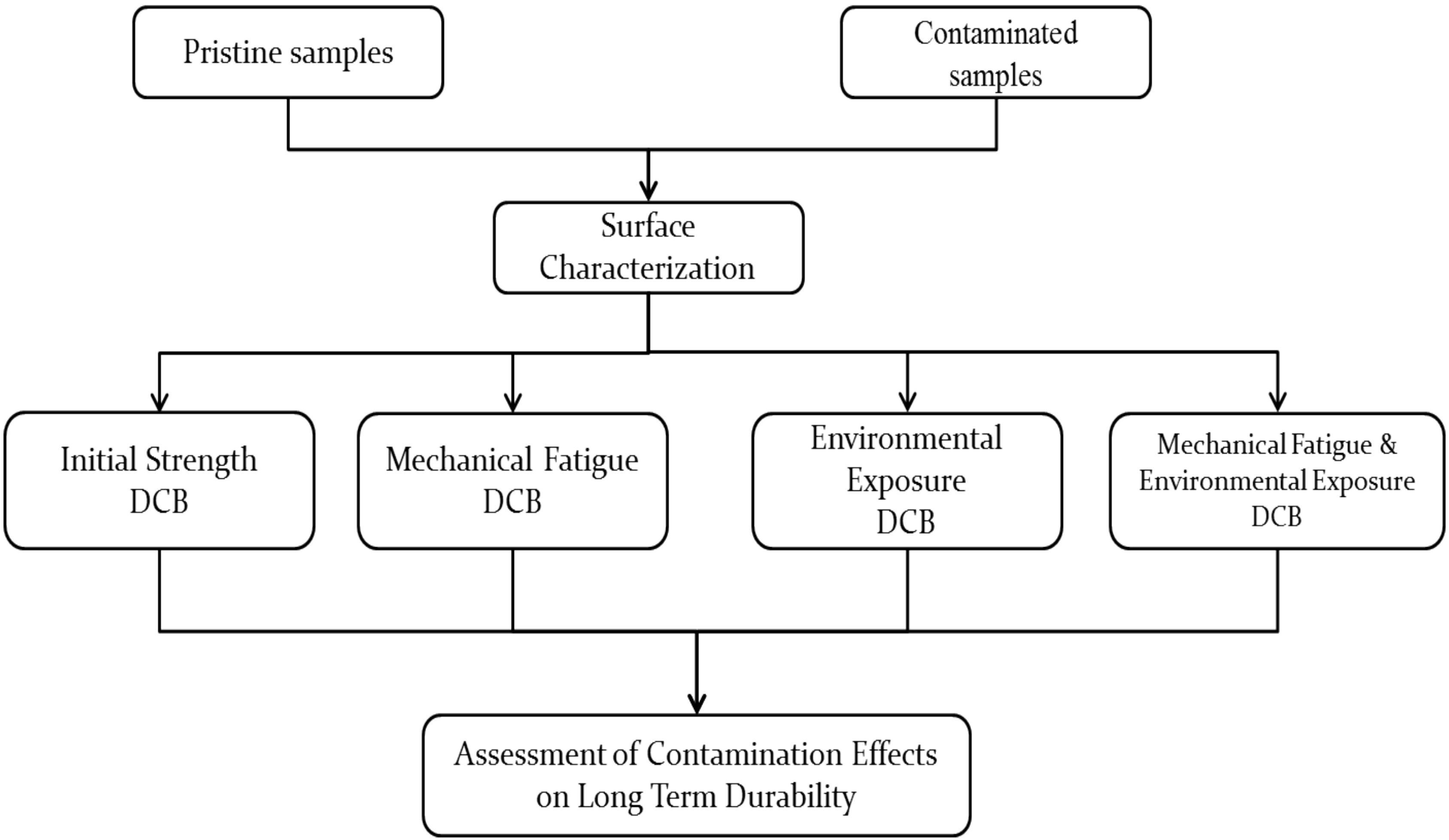
***Motivation and Key Issues***

- Past research has focused on determining/understanding acceptable performance criteria using the initial bond strength of composite bonded systems.
- There is significant interest in assessing the durability of composite bonded joints and the how durability is effected by contamination.

***Objective***

- Develop a process to evaluate the durability of adhesively bonded composite joints
- Investigate **undesirable bonding conditions** by characterizing the initial performance and at various contamination levels
- Characterize the durability performance of the system using the same contamination levels

# Durability Assessment Procedure



# Bonding System Materials

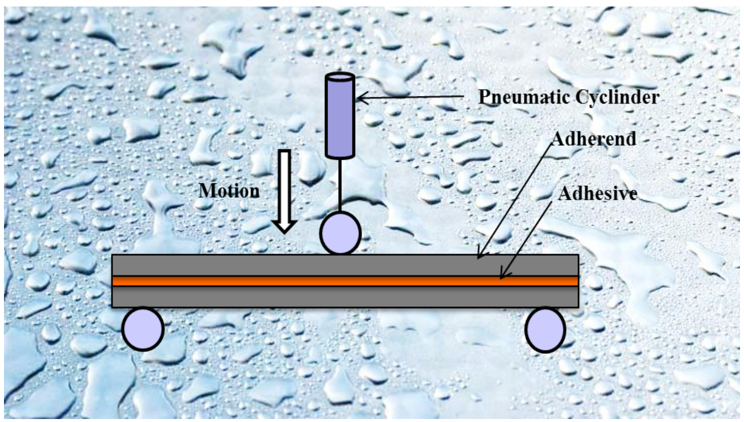
- ***Material type and curing procedure for specimens:*** unidirectional carbon-epoxy system, film adhesive, secondary curing bonding and contaminants.

- ***Materials utilized:***
  - Toray P 2362W-19U-304 T800 Unidirectional Prepreg System (350F cure)
  - 3M AF 555 Structural adhesive film (7.5x2 mills, 350F cure)
  - Precision Fabric polyester peel ply 60001
  - Silicone Spray from CBS Aerosol & Paint, Inc
  - Freekote 700-NC from Henkel Corporation

- ***Specimen Conditioning:***
  - Environmental Chamber : 50°C, 95% RH, for 8 weeks
  - Fatigue Loading: 3 point bending arrangement, 1 inch double amplitude, 2.6 million cycles

# Fatigue Loading Procedure

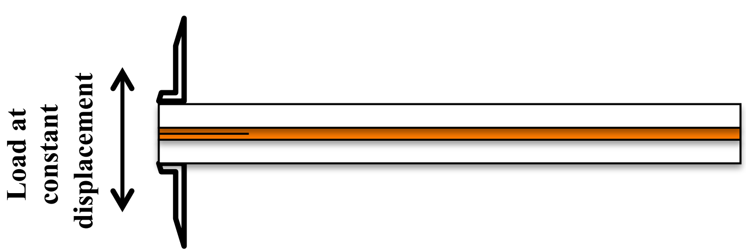
DCB specimens are conditioned by mechanically fatiguing and/or exposure to an accelerated aging environment. **A fatigue structure was manufactured that loads the specimens in three point bending.**




## Advantages:

- Apply uniform shear stress at bondline
- Simple to set up – potential to enclose in an environmental chamber
- Can use DCB (ASTM 5528) or wedge specimens (ASTM 3762)

## Disadvantages:

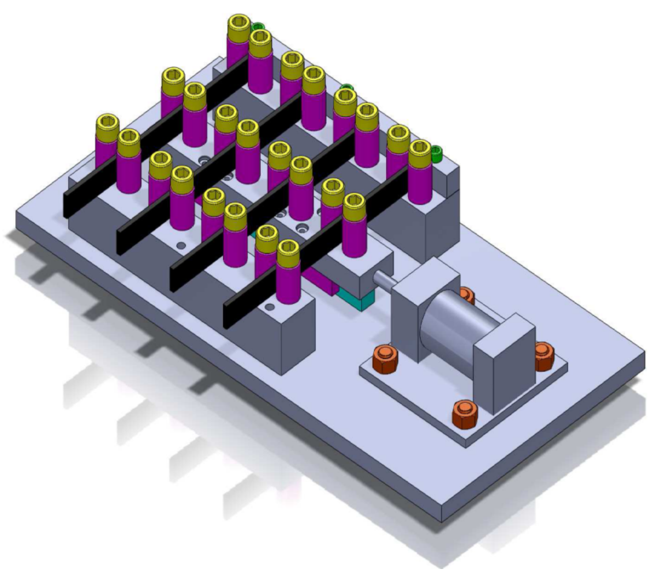



- Specimen geometry needs to be adjusted to  to limit fatigue in adherend/adhesive
- Need to consider surface stress effects resulting from contact points

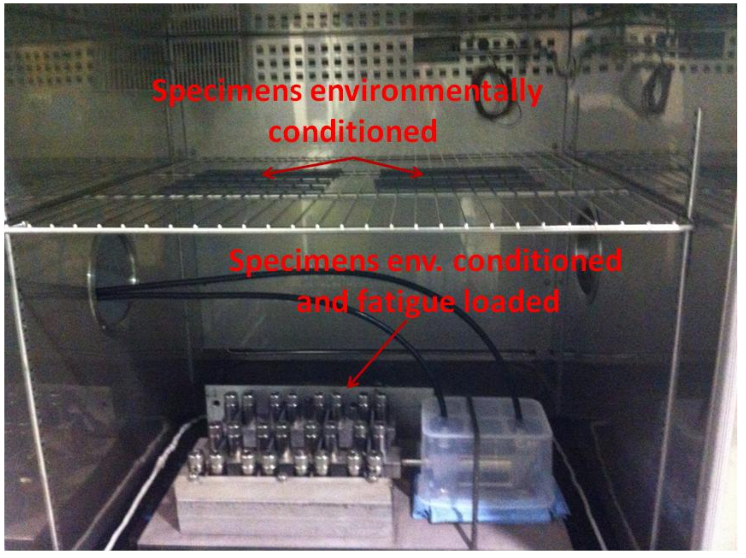
# Accelerated Aging Procedure

**The fatigue fixture can be placed in the environmental chamber to study the combined loading and environmental effects.**

- Manufactured using stainless steel materials
- Center section slides on a ball bearing carriage
- Designed to load up to four 11.5 in specimens with a deflection up to 2 inches DA
- Current stainless steel pneumatic /hydraulic actuator is rated to 400 psi with a 1 inch bore diameter
- Pneumatic controller can operate up to 2 Hz at 150 psi



**Rendering of fatigue fixture**




**Environmental chamber with fatigue fixture**

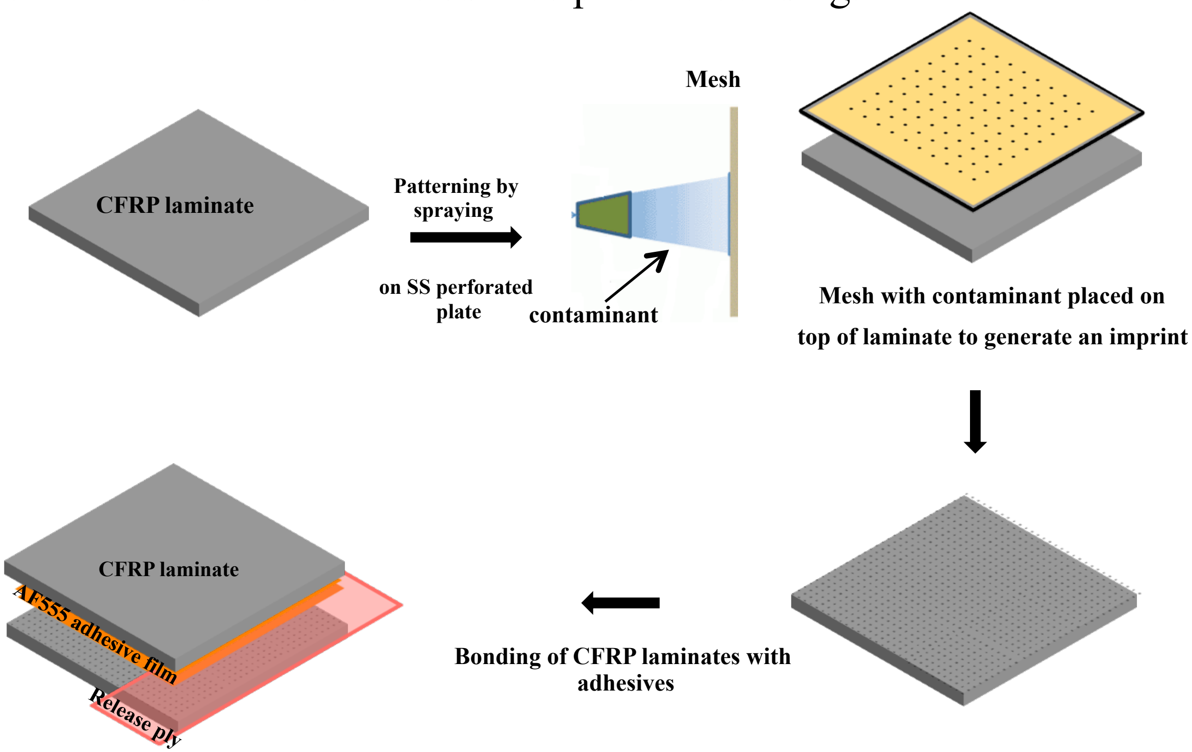
# Contamination Procedure

Undesirable bonding conditions will be used to evaluate how the specimen conditioning can effect durability. Several approaches for contamination are being considered

- Mesh approach
- Stamped approach

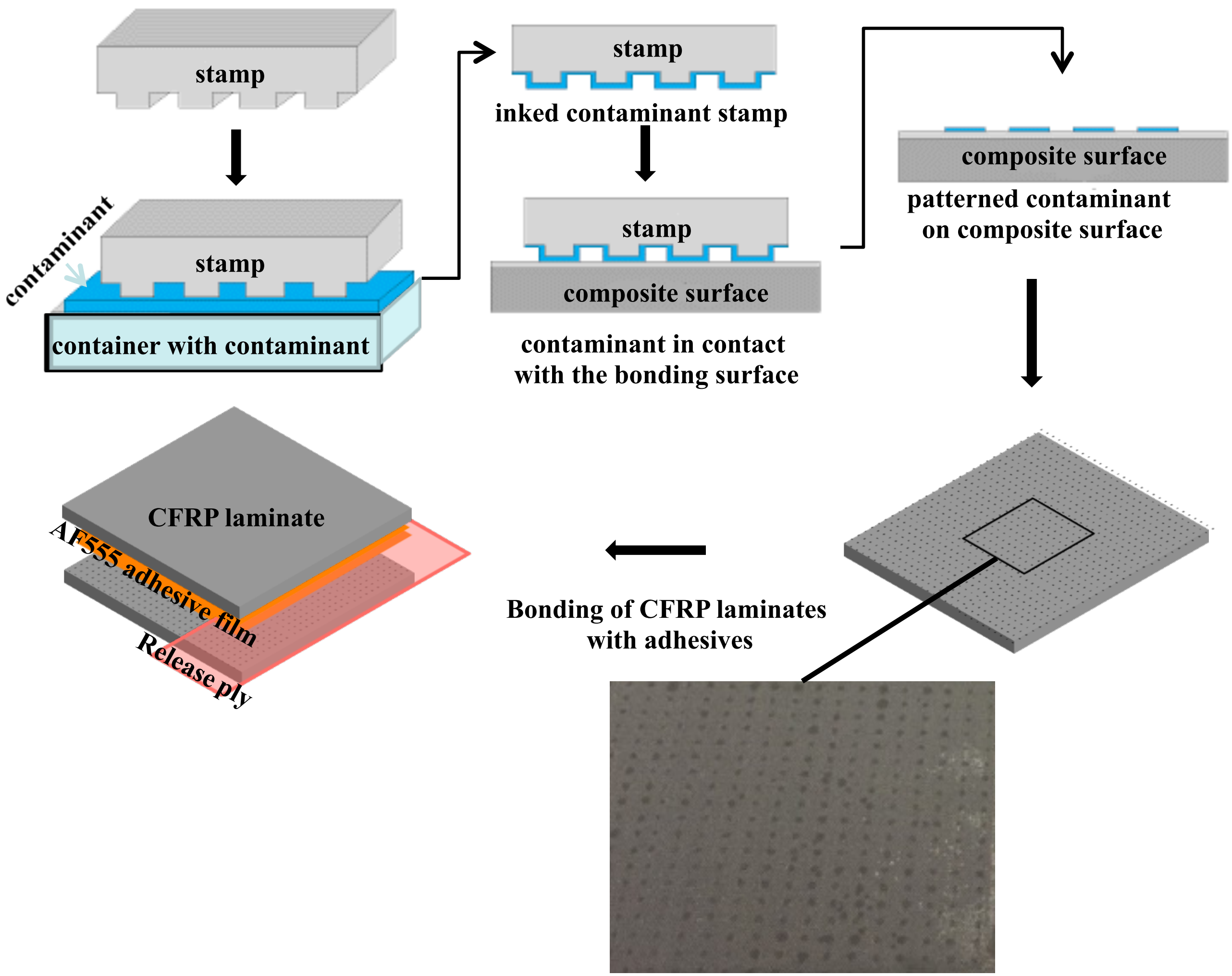
For both cases, the contamination procedure aims to create weak bonds by placing a spatially ordered array of contaminated areas on the surface prior to bonding.

**Mesh Contamination Approach**



# Contamination Procedure

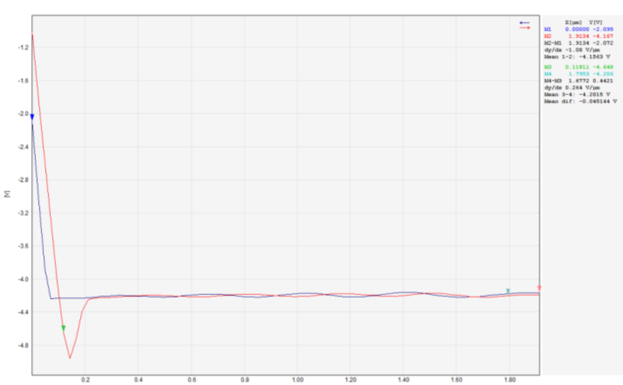
**Stamped Approach**



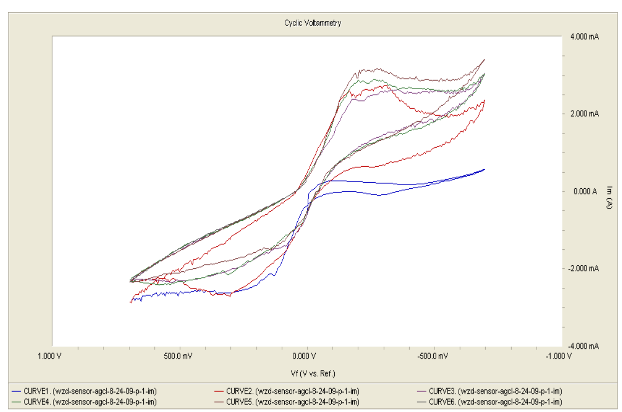
# Surface Characterization Methods

**<u>Pristine and Contaminated Specimens</u>**

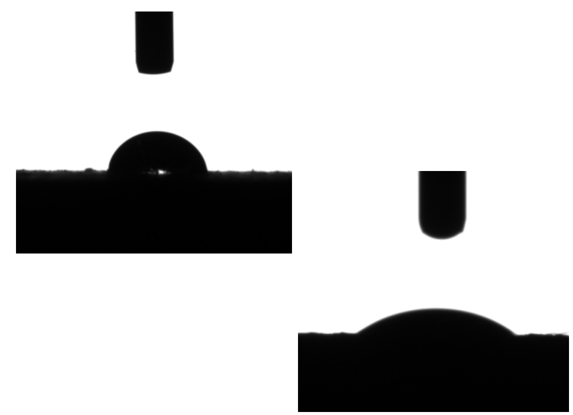
- FTIR data can be used to identify surface molecular bonds – identify contamination.
- Water contact angle - Determines the wetting characteristic behavior of various liquids on the composite surface.
- AFM –can record the attraction/repulsion forces between the AFM probe and the surface. This data is used to generate topography and force volume measurements to quantify changes in adhesion forces.
- Electrochemical Sensor (ECS) - Differences in the measurements reflect levels of electrochemical activity or lack of activity due to contamination.



**Force curve on the peel ply imprint peek obtained**




**CV scans of APCM prepreg and Fiberglast polyester peel ply**



**Contact Angle image of CFRP substrate**

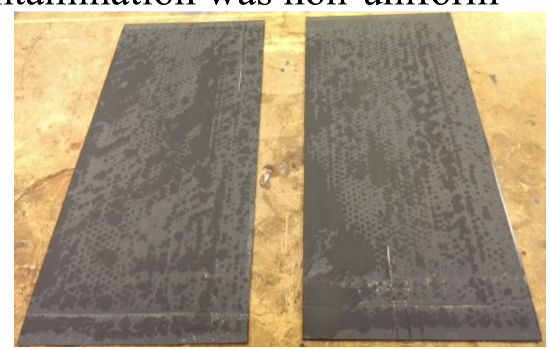
# Surface Characterization Methods

**Fourier transformed infrared spectroscopy**

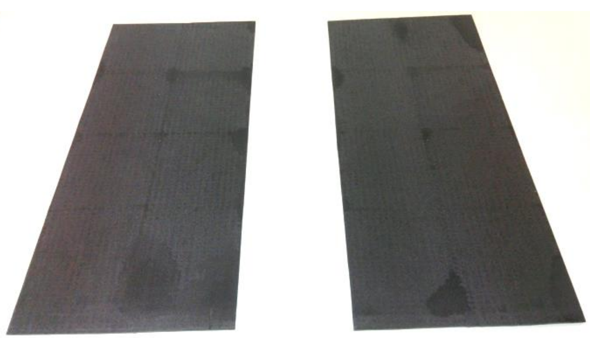
*Aerosol Si Spray Contamination*

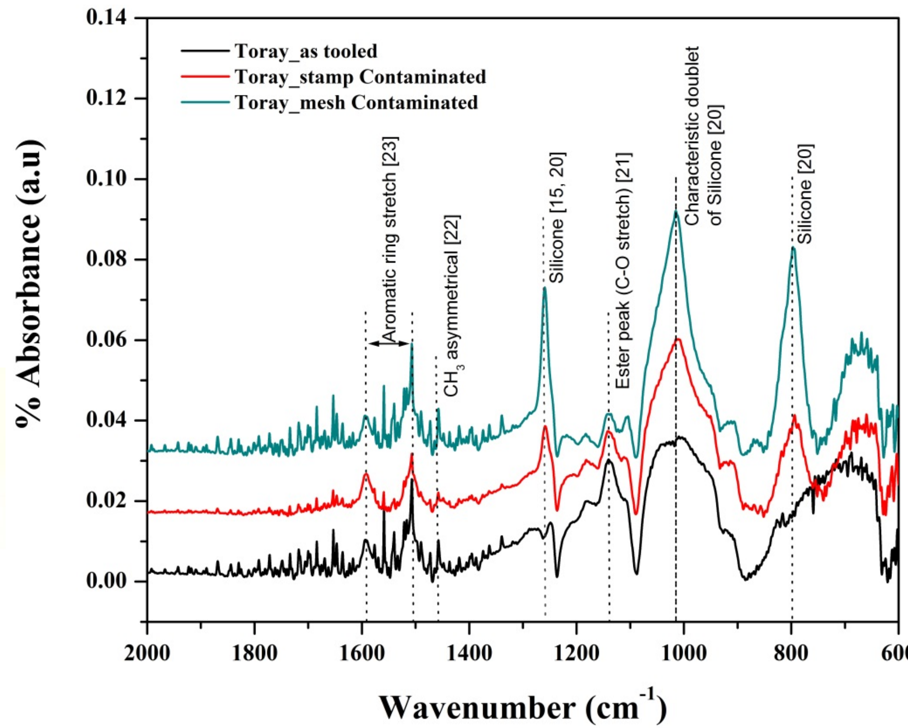
- Baseline specimens showed minimal traces of silicone - contaminated specimens produced sharper peaks
- Mesh specimens had higher silicone peak intensities than stamped specimens but distribution of contamination was non-uniform

**Mesh Approach**



**Stamp Approach**

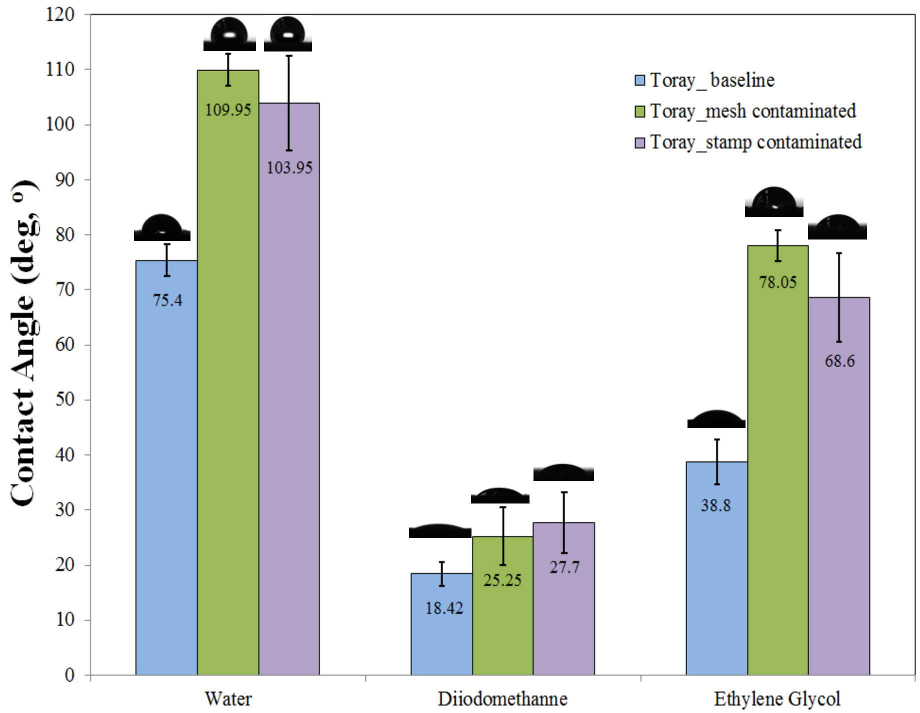


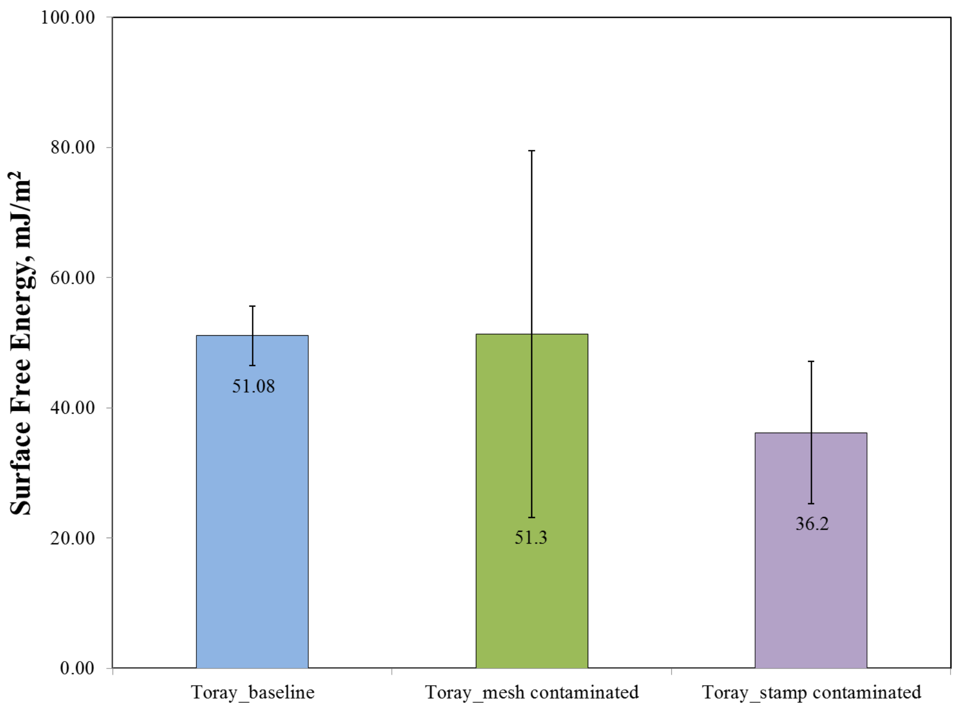


# Surface Characterization Methods

## Contact angle measurements – Surface Energy

3 Fluids - DI water, diiodomethane and ethylene glycol used

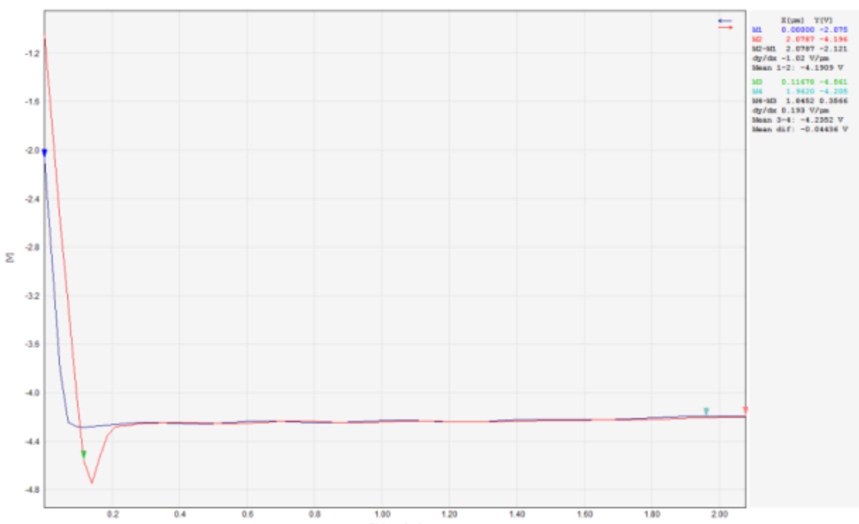


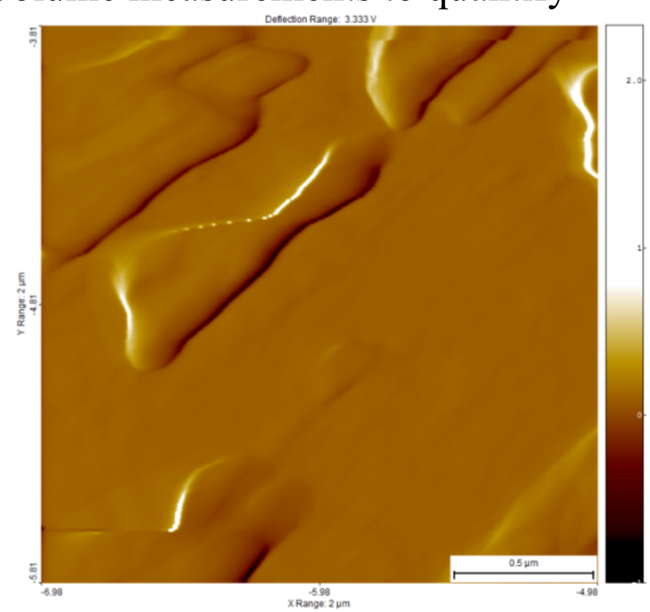
# Surface Characterization Methods

**Atomic force microscopy**

- FIU has conducted research utilizing atomic force microscopy and epoxy-modified probes tip to characterize surfaces prior to bonding.
- AFM can record the attraction/ repulsion forces between the AFM probe and the surface.
- AFM data is used to generate topography and force volume measurements to quantify changes in adhesion forces.




**Typical force deflection curve**



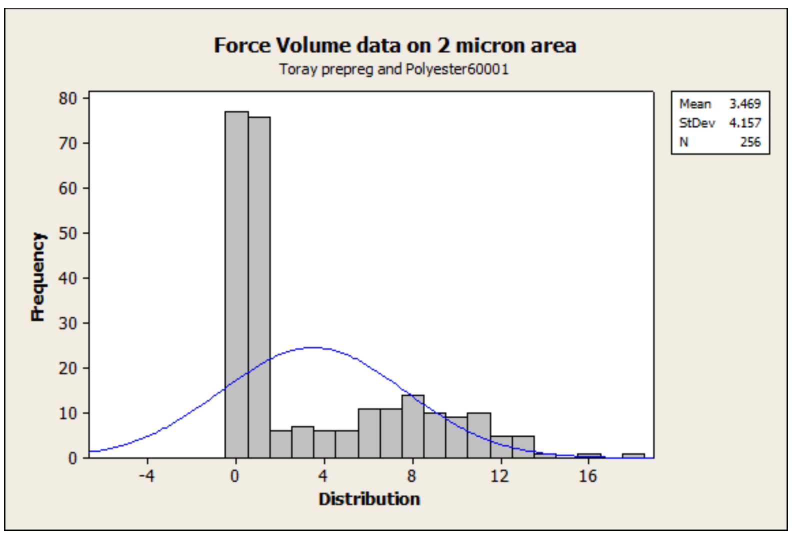

**Topography image of  peel ply imprint peek**

# Surface Characterization Methods

**Atomic force microscopy**

- A 16 x16 grid is used to generate 256 adhesion force measurements using a force volume approach

- Force volume data was collected in a controlled environment (~0% humidity) and in ambient air




**Histogram of adhesion force measurements**

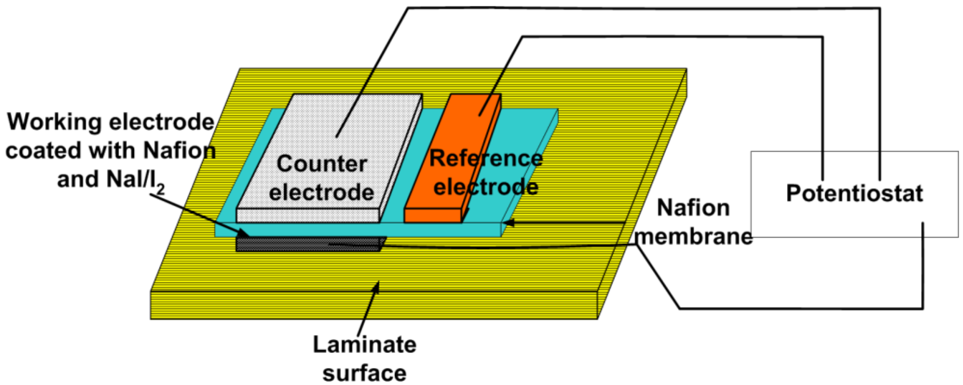
| ***AFM Force-Volume data*** | ***Detach Force (nN)*** | |
|---|---|---|
| | *Ambient Air* | *Controlled Environment* |
| ***Mean*** | *1.35* | *3.47* |
| ***Standard Deviation*** | *0.98* | *4.16* |
| ***Maximum*** | *6.03* | *18.44* |
| ***Minimum*** | *0.02* | *0.20* |

**Adhesion force measurement data**

**(only baseline)**

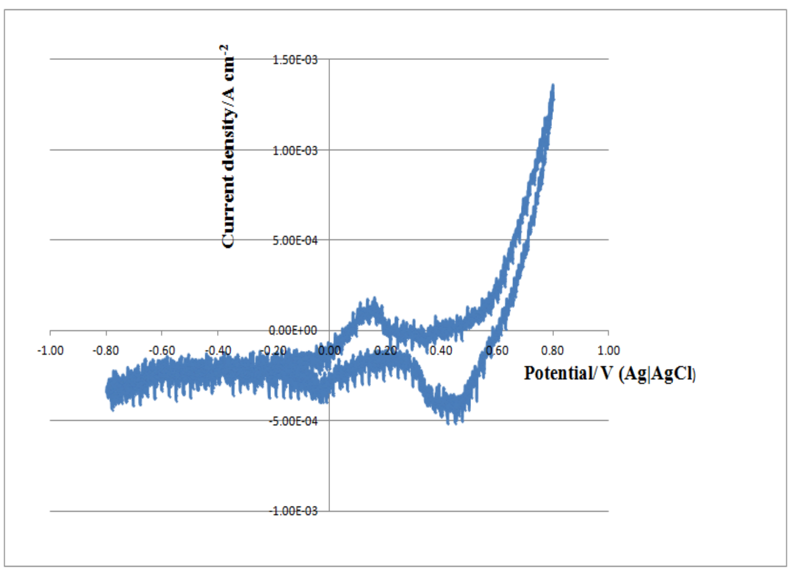
# Surface Characterization Methods

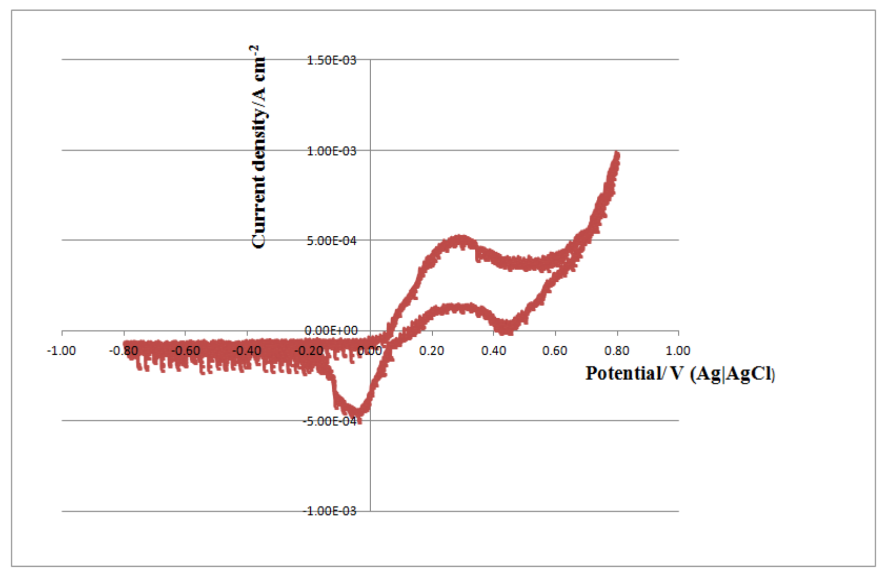
**Electrochemical sensor measurements**



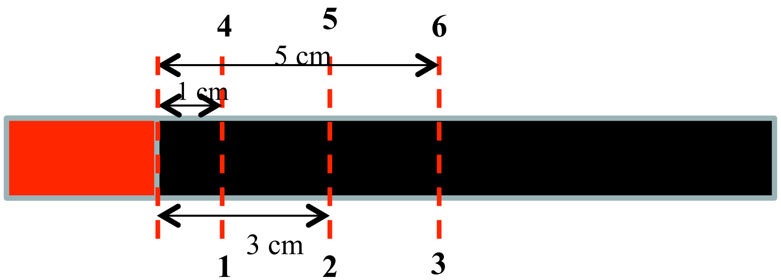

**Schematic of the solid-state ECS**

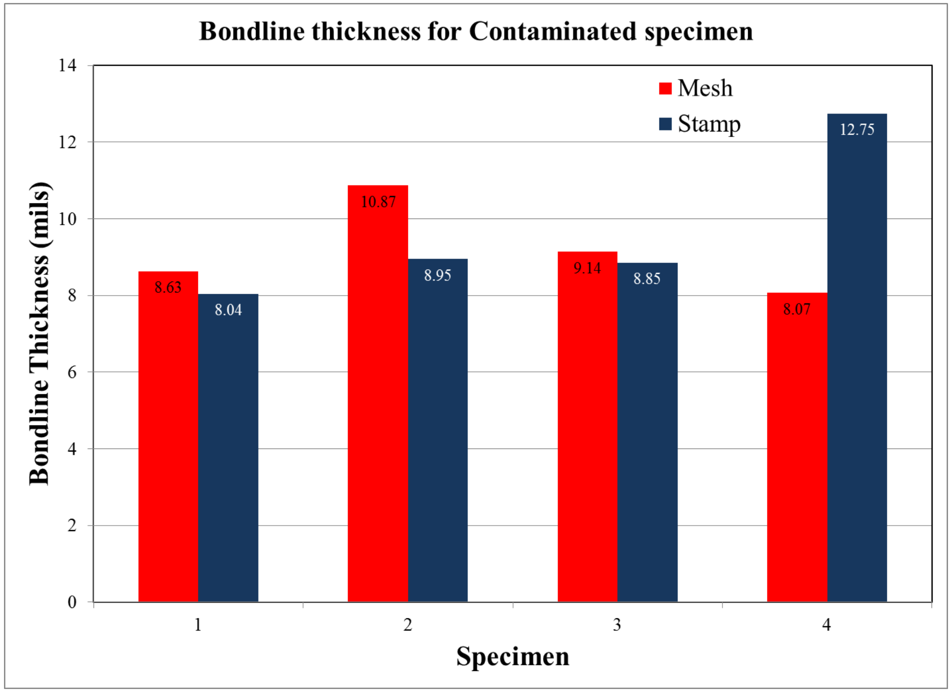
- Measure electrochemical activity on surface by varying the potential and evaluating the current response – cyclic voltammetry
- CV peaks can reflects variations in electrochemical activity and potential contamination



# Bondline Thickness Measurements

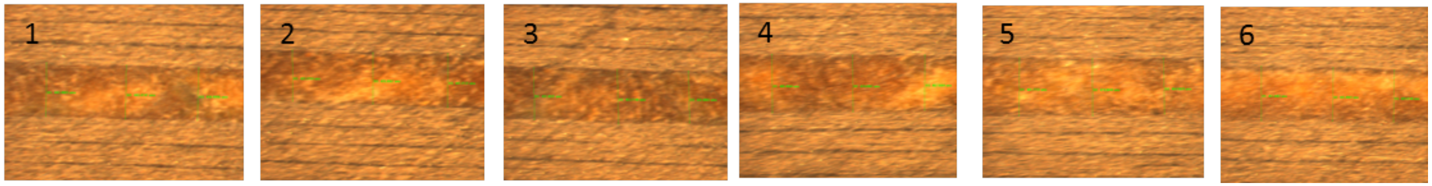


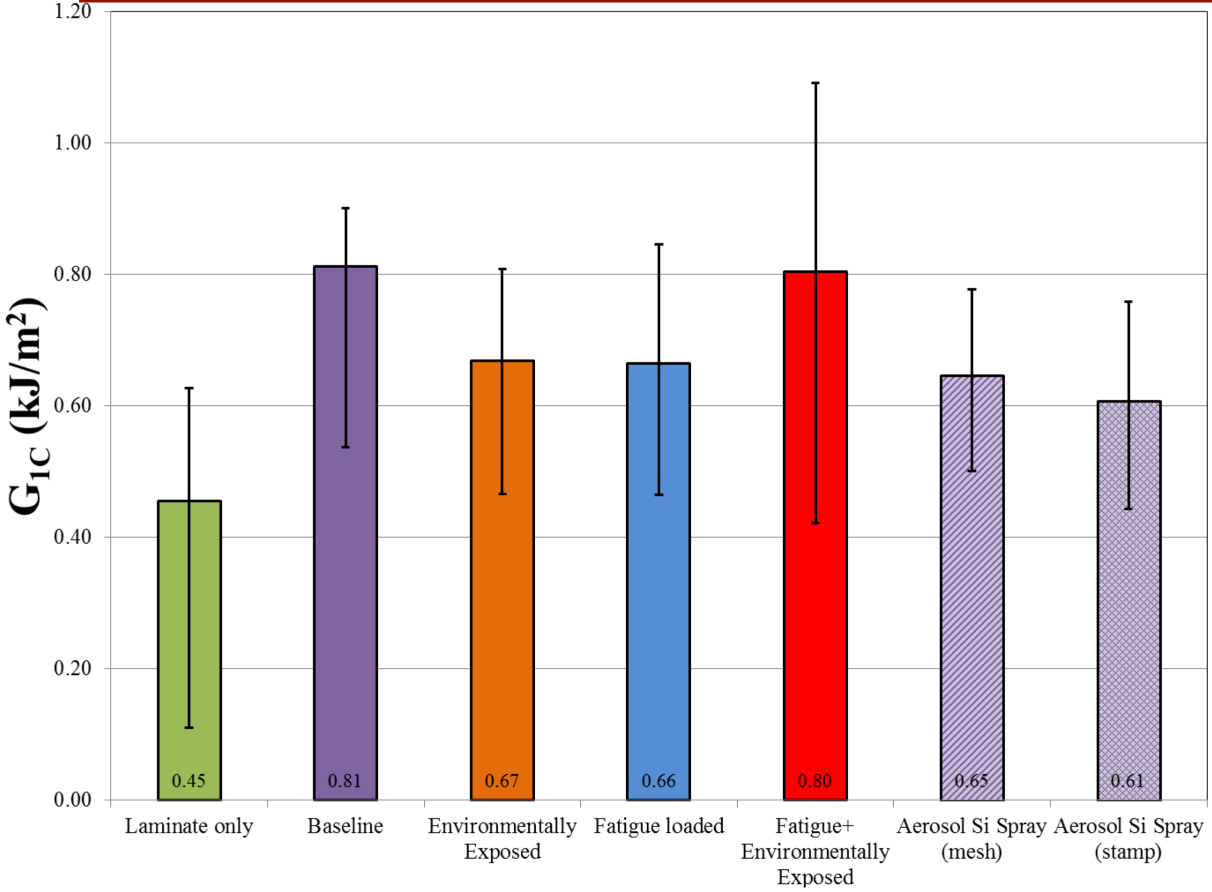

**Avg. Bondline (baseline)- 9.28 mils**

**Avg. Bondline (mesh)- 8.60 mils**

**Avg. Bondline (stamp)- 9.65 mils**



# Fracture Toughness Testing - DCB

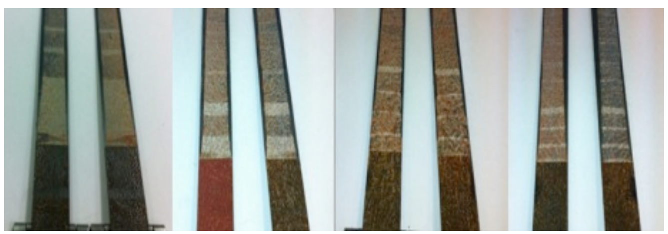



| $G_{IC}$ | ***Average*** (in-lb)/in² | ***Max*** (in-lb)/in² | ***Min*** (in-lb)/in² |
|---|---|---|---|
| *Laminate only* | *0.45* | *0.63* | *0.11* |
| *Baseline* | *0.81* | *0.90* | *0.54* |
| *Environmentally Exposed* | *0.67* | *0.81* | *0.47* |
| *Fatigue loaded* | *0.66* | *0.85* | *0.46* |
| *Fatigue+ Environmentally Exposed* | *0.80* | *1.09* | *0.42* |
| *Contaminated-Mesh Stamp* | *0.65* | *0.78* | *0.50* |
| *Contaminated-Rubber Stamp* | *0.61* | *0.76* | *0.44* |

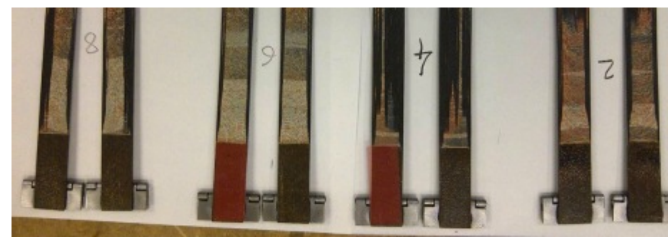
# Modes of Failure

a)

b)



c)



d)

e)

f)

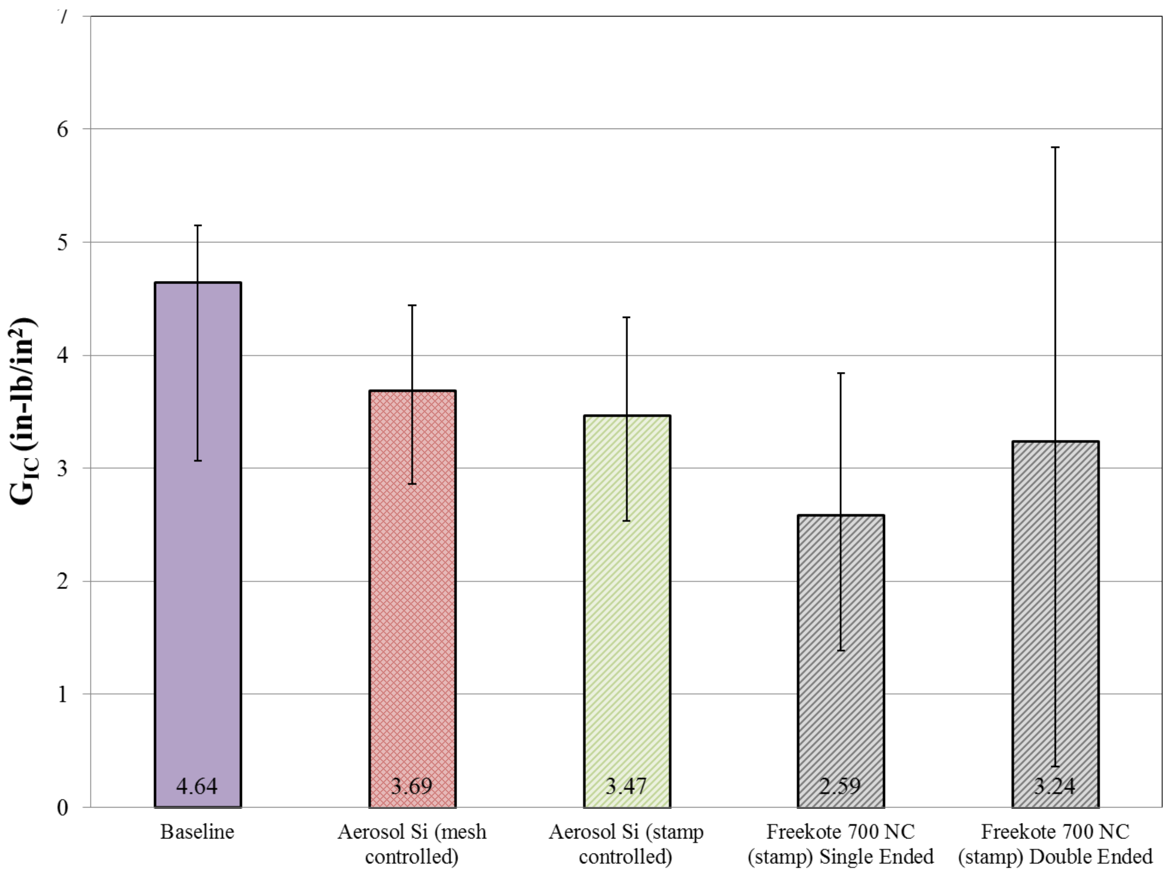
A - pristine B - environmentally exposed C - fatigued D - environmental exposed and fatigued

<u>Contaminated Specimens (Aerosol)</u>

E –mesh controlled F - stamp controlled

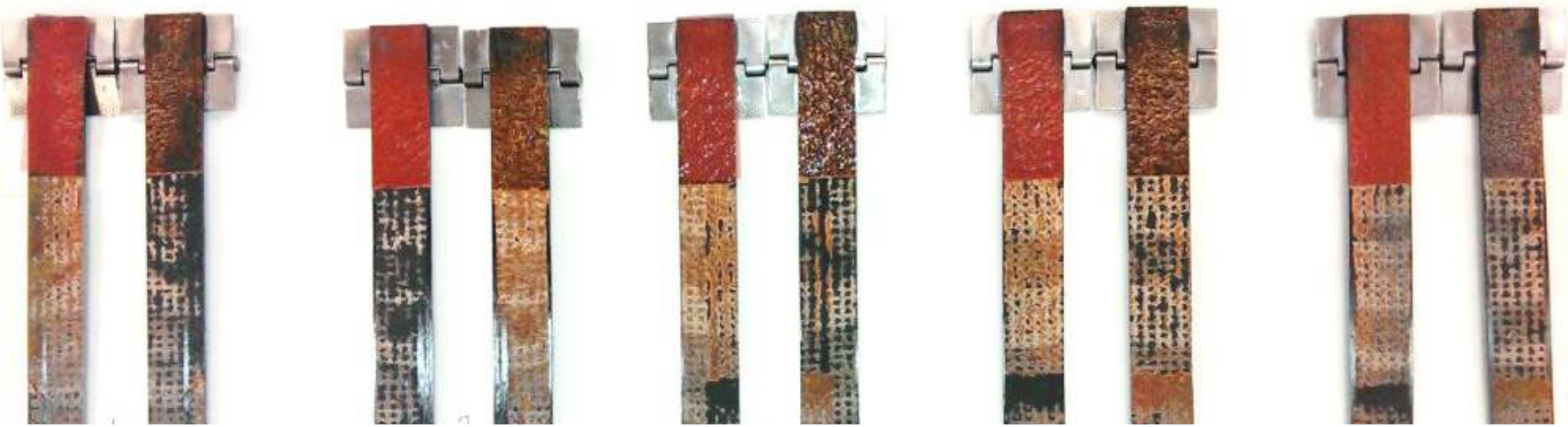
# Alternate Contaminant- Freekote 700 NC

## Comparison of contaminants




| $G_{IC}$ | *Average* (in-lb)/in² | *Max* (in-lb)/in² | *Min* (in-lb)/in² |
|---|---|---|---|
| *Laminate only* | *4.64* | *5.15* | *3.07* |
| *Contaminated-Aerosol-Mesh* | *3.69* | *4.44* | *2.86* |
| *Contaminated-Aerosol-stamp* | *3.47* | *4.33* | *2.53* |
| *Contaminated-Freekote-stamp_single ended* | *2.59* | *3.84* | *1.39* |
| *Contaminated-Freekote-stamp_double ended* | *3.24* | *5.84* | *0.36* |

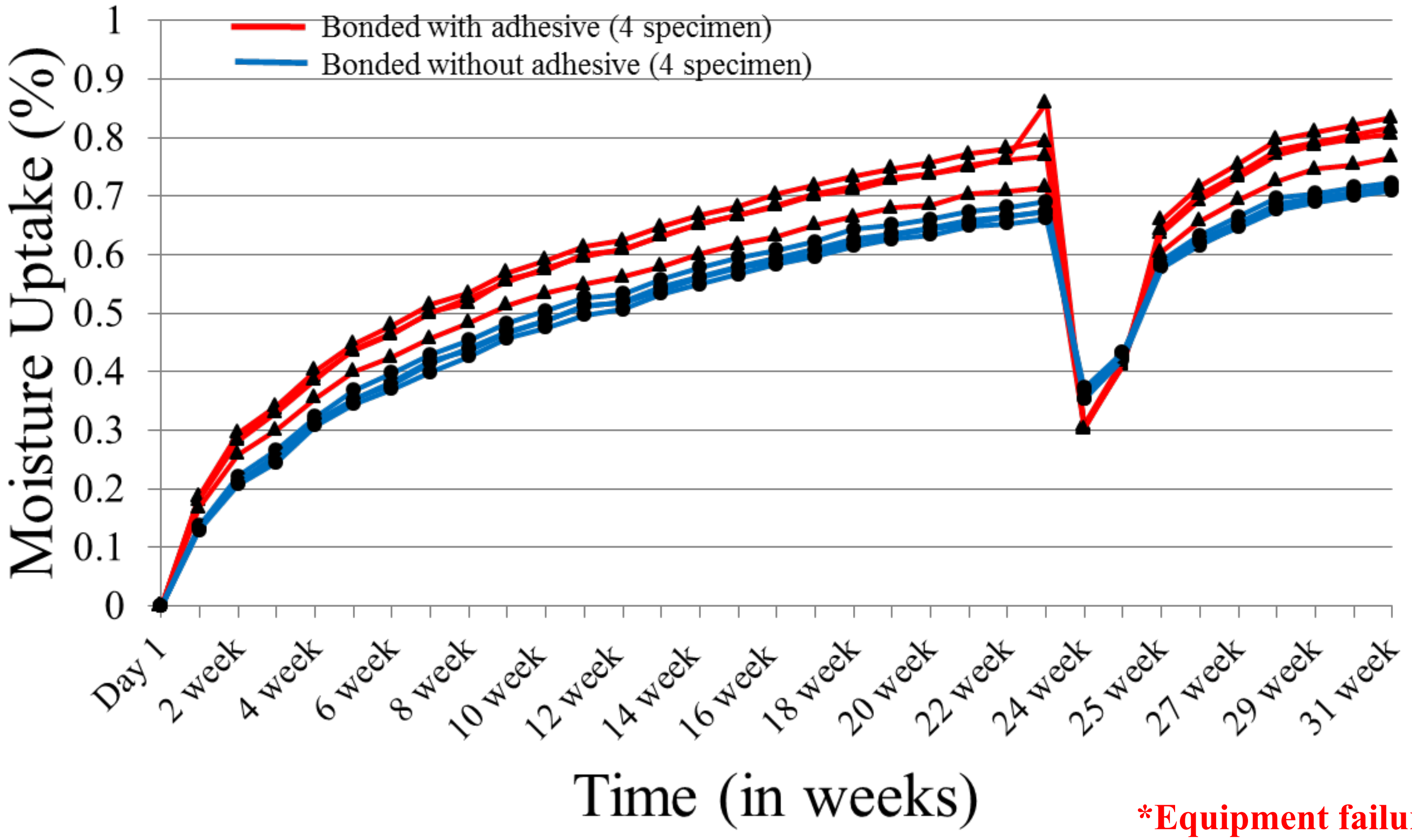
# Failure Mode of Contaminated Specimens



**DCB Specimens contaminated with Freekote 700-NC**

# Weight Measurements

**Moisture uptake analysis – determine saturation limit**




*Equipment failure @ week 23

# Conclusions

- A general contamination procedure has been developed to evaluate the effect of contamination on adhesively bonded joints.
- Durability assessment by conditioning of specimens using a 3 point bending fixture for mechanical fatiguing in air and in environmental chamber.
- Surface characterization (Contact Angle and FTIR) showed a different surface chemistry on contaminated specimens when compared to pristine specimens.
- A significant degradation in fracture toughness was observed for the Freekote contaminated specimens when compared with baseline specimens.
- Adhesion failure modes were observed with the Freekote contamination. Mixed cohesive and interlaminar failure modes were observed in others specimens.
- Moisture uptake curve continues to asymptotically increase - equilibrium saturation limit will continue to be monitored.

# Path Forward

**Future Work:**

- Characterize contaminated surfaces prior to bonding (AFM, ECS)
- Repeat conditioning on contaminated specimens (environmentally aging and mechanical fatiguing)
- **Measure bond degradation of conditioned contaminated specimens – DCB testing**
  - **Repeat contamination procedure to provide larger contamination areas**

**Benefit to Aviation:**

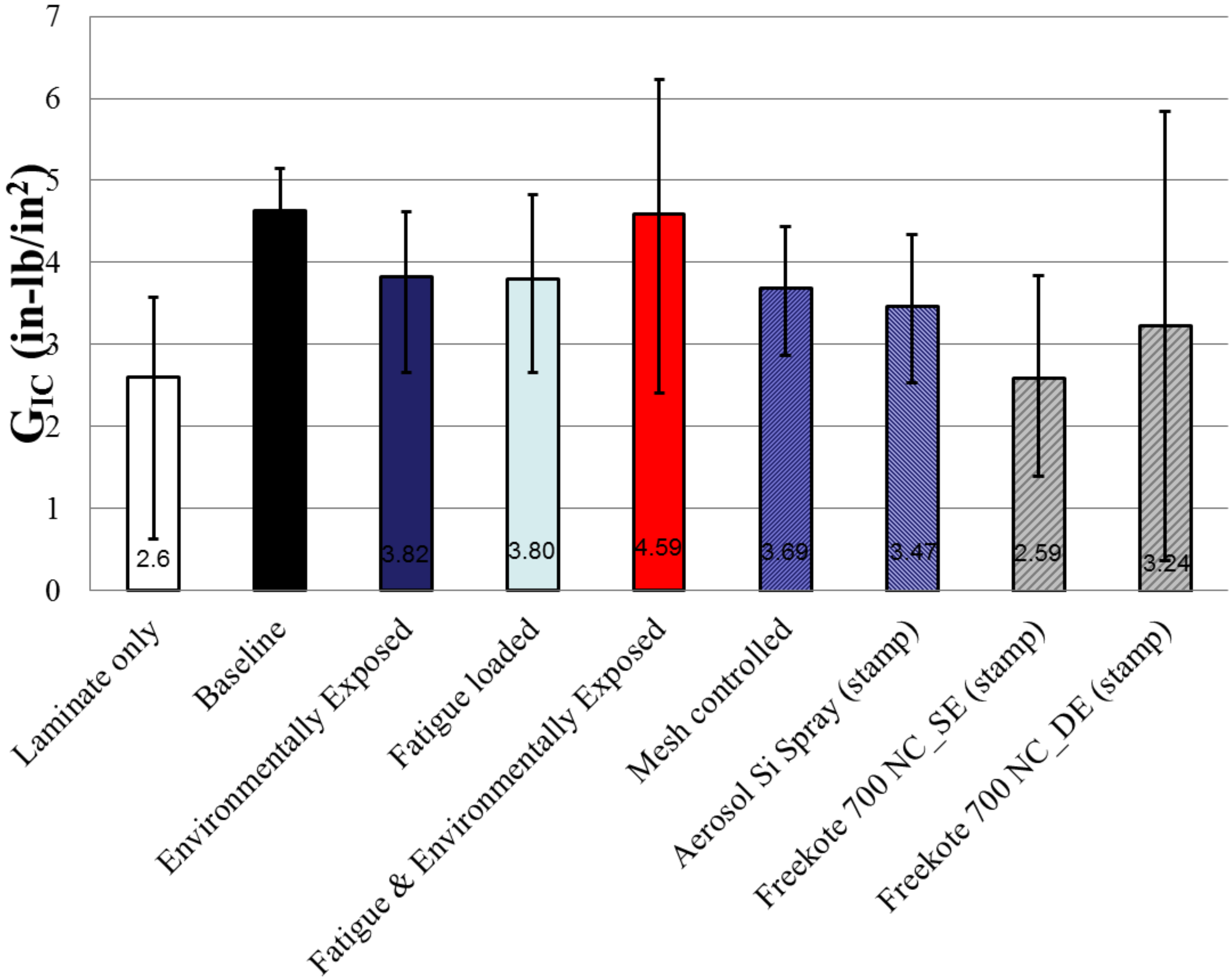
- Better understanding of durability assessment for adhesively bonded composite joints.
- Assisting in the development of bonding quality assurance procedures.

Questions?

# Weight Gain from Contamination

| Contaminant Deposition - Aerosol | | | | | |
|---|---|---|---|---|---|
| | | | Before (gms) | After (gms) | Gain % |
| Aerosol | Aerosol Si Spray (stamp) | Laminate 1 | 123.04 | 123.05 | 0.008 |
| | | Laminate 2 | 124.18 | 124.24 | 0.048 |
| | Aerosol Si Spray (mesh) | Laminate 1 | 123.59 | 123.63 | 0.032 |
| | | Laminate 2 | 122.52 | 122.54 | 0.016 |

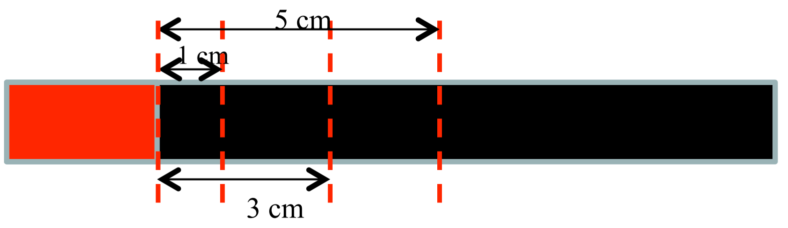
| Contaminant Deposition – Freekote (Stamp Approach) | | | | | |
|---|---|---|---|---|---|
| | | | Before (gms) | After (gms) | Gain % |
| Freekote | Single Ended | Laminate 1 | 124.97 | 125.04 | 0.056 |
| | | Laminate 2 | 127.48 | 127.53 | 0.039 |
| | Double Ended | Laminate 1 | 126.4 | 126.45 | 0.039 |
| | | Laminate 2 | 127.3 | 127.34 | 0.031 |



$G_{IC}$ (in-lb/in$^2$)
0
1
2
3
4
5
6
7
2.6
3.82
3.80
4.59
3.69
3.47
2.59
3.24
Laminate only
Baseline
Environmentally Exposed
Fatigue loaded
Fatigue & Environmentally Exposed
Mesh controlled
Aerosol Si Spray (stamp)
Freekote 700 NC_SE (stamp)
Freekote 700 NC_DE (stamp)

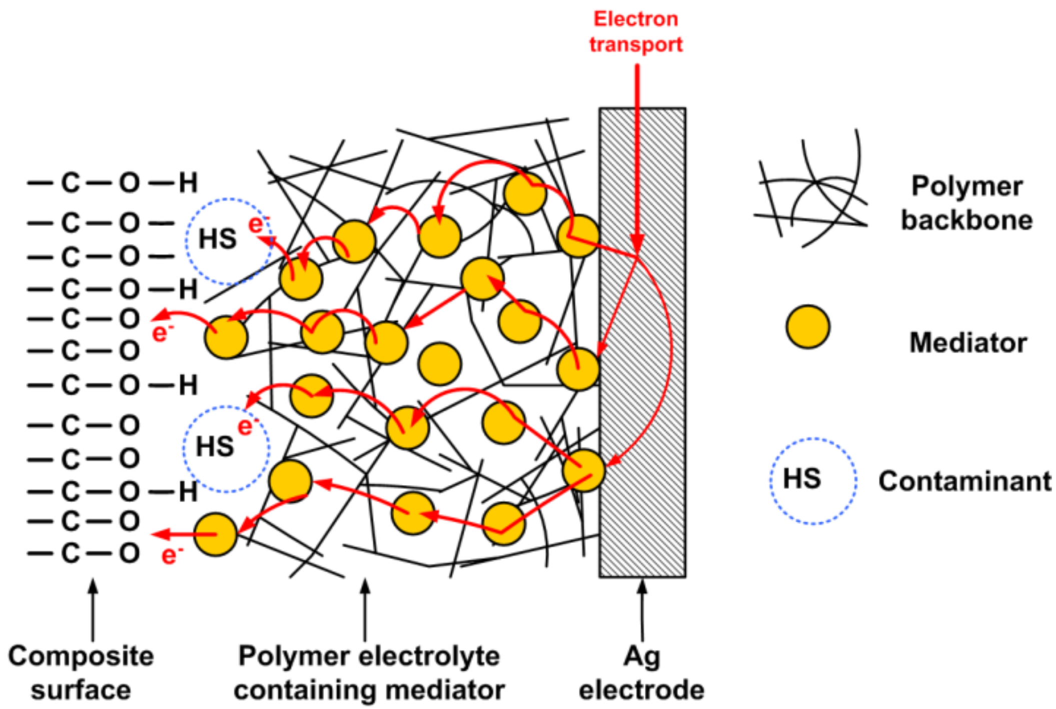
# Specimen Description

**Single Ended DCB specimen**




**Double Ended DCB specimen**

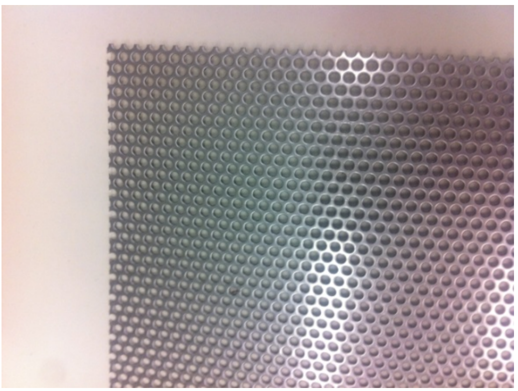
# ECS Operational principle

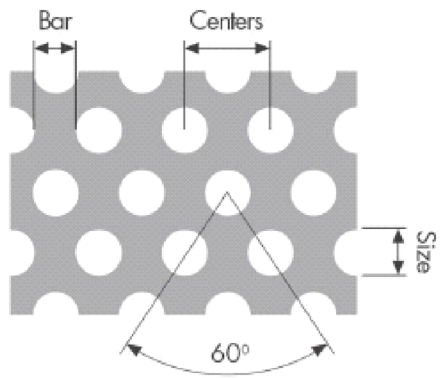



Operation principle of the solid-state ECS

# Contamination Procedure

**Stainless Steel Mesh**

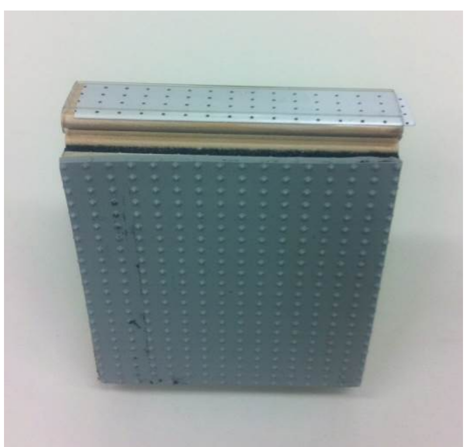





Dimensions

| Dimension Name | Value |
|---|---|
| Thickness | 0.036" |
| Hole Size | 0.125" |
| Stagger | 0.1875" |
| Open Area | 40% |

**Rubber Stamp**



**3 in X 3 in equally spaced dots**

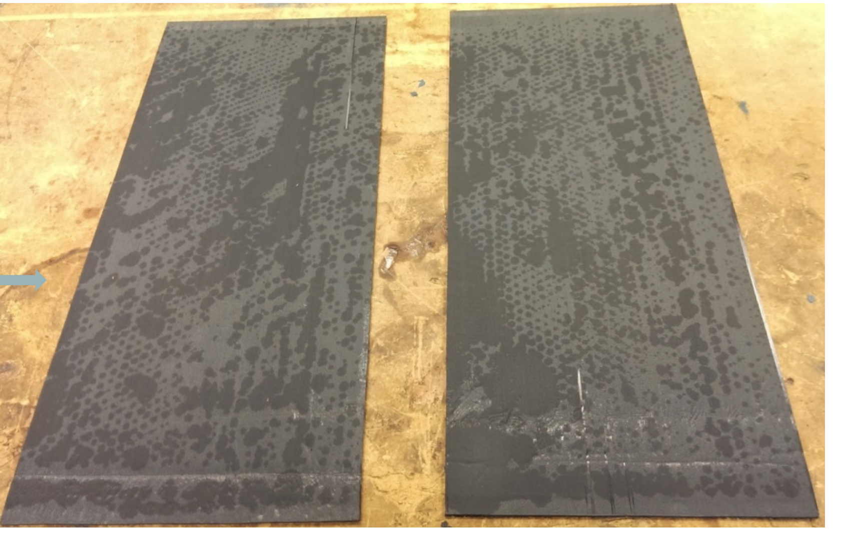
**(New Times Roman, font size 12)**

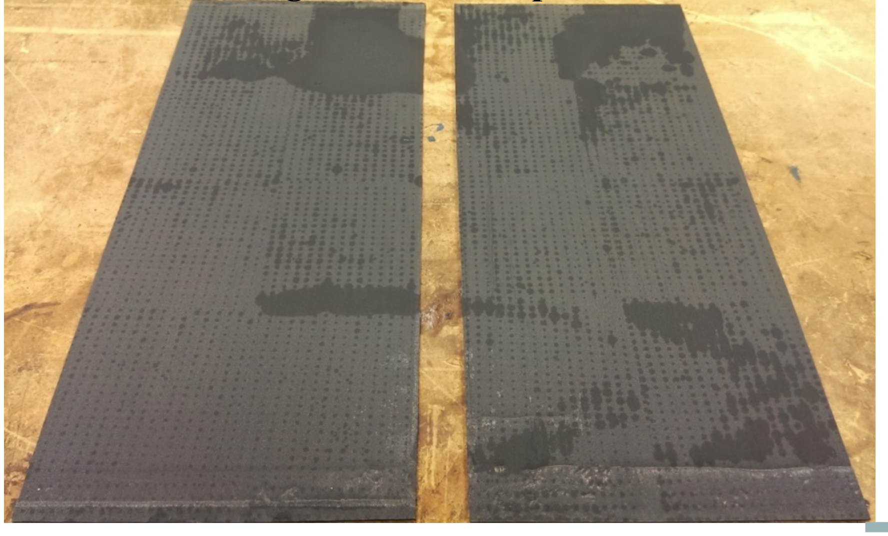
# Contaminated Laminates Prior to Bonding

**Mesh Stamp Contamination**

**Contaminant- 100% pure Silicon spray (from CBS Aerosol & Paint, Inc)**

**Directions: Sprayed on the mesh initially and then mesh placed on the laminate surface to generate the imprint.**





**Rubber Stamp Contamination**

**Contaminant- 100% pure Silicon spray (from CBS Aerosol & Paint, Inc)**

**Directions: Rubber stamp with the dotted pattern carved- Stamp is pressed on Silicon medium and transferred onto the laminate**

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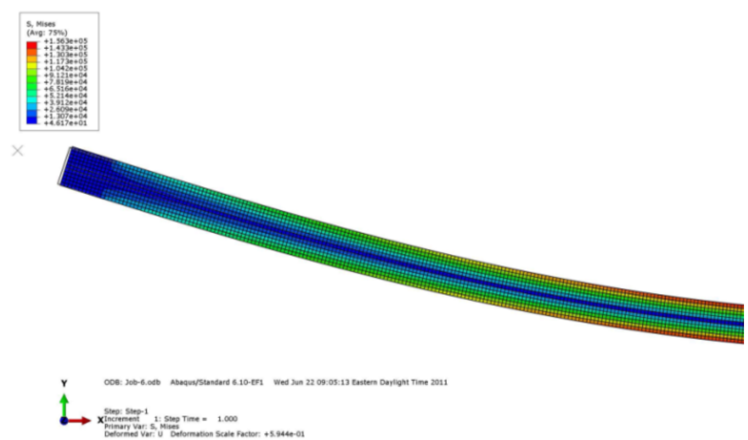
## SECTION 2: INGREDIENTS

| Ingredient | C.A.S. No. | % by Wt |
|---|---|---|
| ISOBUTANE | 75-28-5 | 70 - 80 |
| HEPTANE | 142-82-5 | 15 - 25 |
| POLY(DIMETHYLSILOXANE) | 63148-62-9 | 3 - 7 |

| Max Stress (psi) | Avg. Life (Cycles) |
|---|---|
| 4500 | $1.58 \times 10^4$ |
| 4000 | $5.28 \times 10^4$ |
| 3500 | $4.75 \times 10^5$ |
| 3000 | $2.67 \times 10^6$ |
| 2200 | $1.03 \times 10^7$ + (No failure) |

| # Plies | Thickness (inches) | Force required by piston (lb) | Stress at Surface (ksi) |
|---|---|---|---|
| 16 | 0.120 | 240 | 225 |
| 18 | 0.135 | 270 | 200 |
| 20 | 0.150 | 300 | 180 |
| 22 | 0.165 | 330 | 164 |
| 24 | 0.180 | 360 | 150 |



**Selected laminate configuration:**

- Specimen dimensions: 11.5 in long x 1 in wide
- 20 ply unidirectional laminate (**0.15 thick** + adhesive)