INTRODUCING INTERFACIAL BOND FAILURE AT THE ELASTOMER-PRIMER INTERFACE USING TAILORED INFRARED-SONICATION HYBRID TECHNOLOGY

WP18-1347

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Molekule Inc

9/22/2019

SERDP
DOD ▪ EPA ▪ DOE

Project Team

- **Technical Lead (Molekule) : Dr. Japreet Dhau, Ph.D., MBA**
- Co-performer (Molekule) : Dr. Chatura Wickramratne, Ph.D.
- PI (Florida Polytechnic University) : Nicole Tardif, Ph.D.
- Co-performer (AFRL) : Alan Fletcher

Background

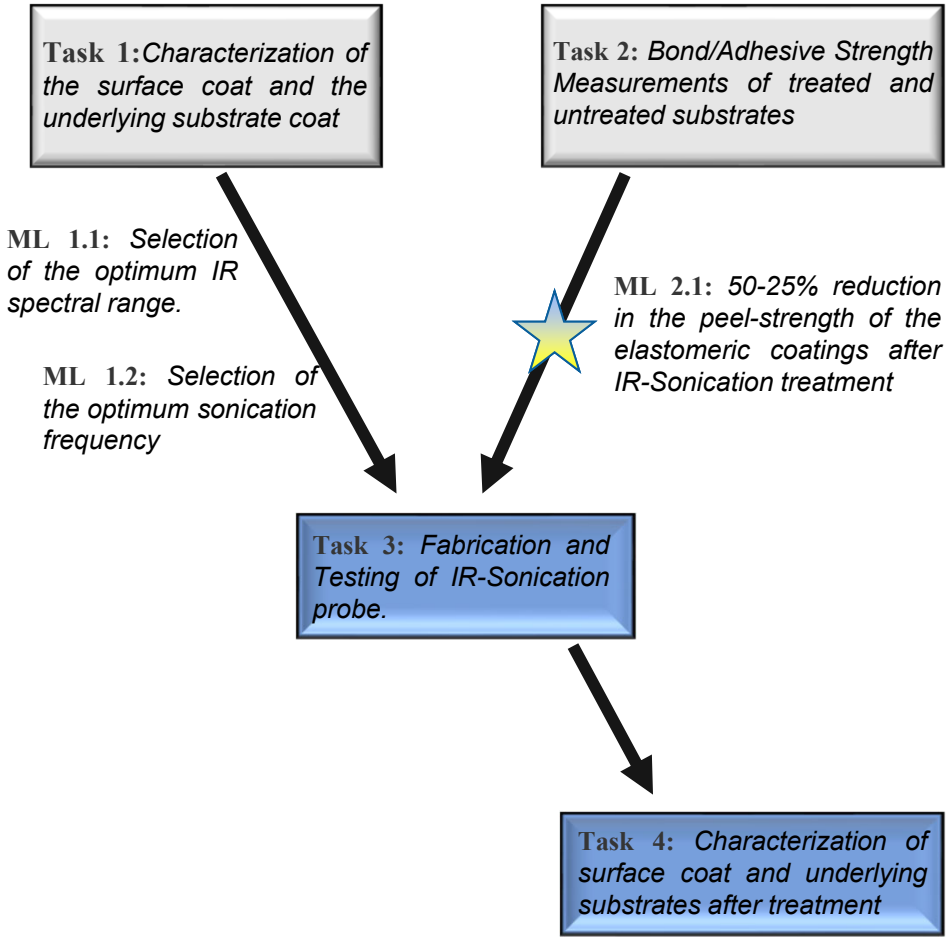
- The project was initiated in August 2018
- Addressed the SERDP's goal to develop a large-scale non-chemical, non-media removal process for thick, elastomeric coatings and treatments, used on Department of Defense (DOD) Weapon Systems.
- Difficulties in removing specialty coatings in an environmentally friendly manner

Technical Objectives

- Induce interfacial bond failure between the coating and the underlying substrate while not damaging the substrate surface

- Developing an innovative and cost-effective ergonomic Infrared-Sonication hybrid technology for removing thick, elastomeric specialty coatings

Technical Approach

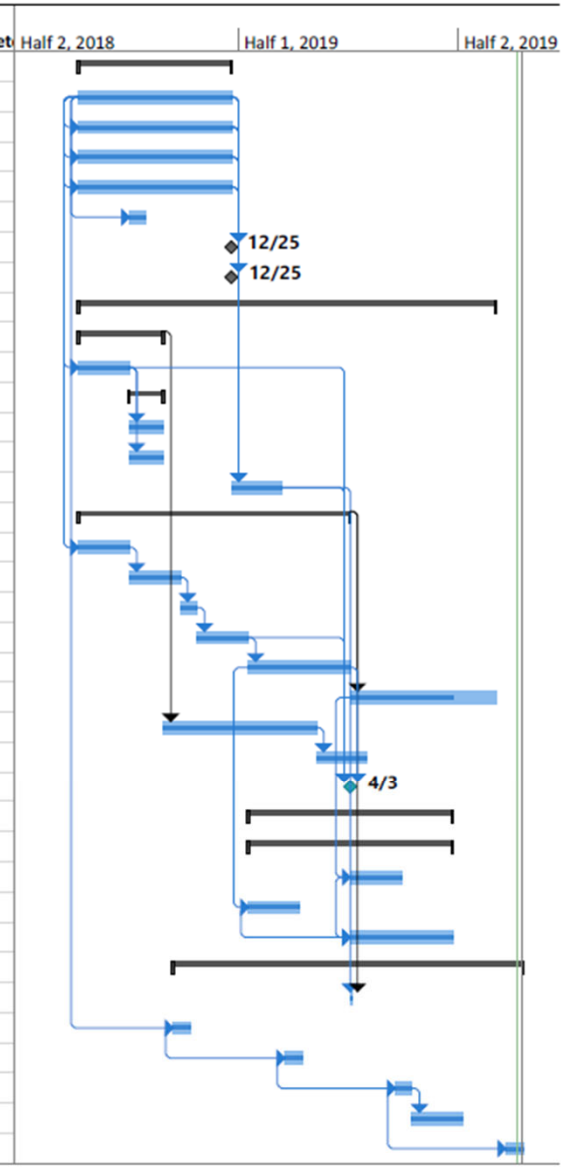


- One of the key significant millstone of **50-25%** reduction in the peel-strength of the elastomeric coatings after IR-Sonication treatment (Milestone 2.1) was achieved with far better results than we initially proposed.

- The average peel strength reduction for all the coatings types and thickness tested were in the **75-50%** range.

Project Plan and Timeline

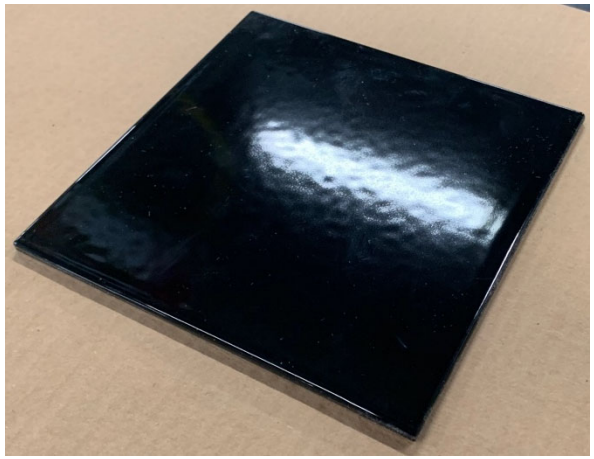
ID	Task Name	Duration	Start	Finish	% Complet
1	**1 Characterization of the surface coat and the underlying substrate**	**90 days**	**Wed 8/22/18**	**Tue 12/25/18**	**100%**
2	1.1 FTIR and Specular reflectance for polyurethane-filler composite	90 days	Wed 8/22/18	Tue 12/25/18	100%
3	1.2 FTIR and Specular reflectance for the underlying substrates	90 days	Wed 8/22/18	Tue 12/25/18	100%
4	1.3 DSC analysis of surface coat polyurethane-filler composite	90 days	Wed 8/22/18	Tue 12/25/18	100%
5	1.4 DSC analysis of underlying substrates	90 days	Wed 8/22/18	Tue 12/25/18	100%
6	1.5 XPS Analysis of the Coating	10 days	Wed 10/3/18	Tue 10/16/18	100%
7	1.6 M 1.1 Selectionof the optimum IR range	0 days	Tue 12/25/18	Tue 12/25/18	100%
8	1.7 M 1.2 Selection of the sonication range	0 days	Tue 12/25/18	Tue 12/25/18	100%
9	**2 Bond Strength Measurements of treated and untreated substrates**	**246 days**	**Wed 8/22/18**	**Wed 7/31/19**	**100%**
10	**2.1 Testing bond strength of samples before treatment**	**50 days**	**Wed 8/22/18**	**Tue 10/30/18**	**100%**
11	2.1.1 Identifying Testing Standards for Coatings/Adhesives	30 days	Wed 8/22/18	Tue 10/2/18	100%
12	**2.1.2 Developing In-house testing setup**	**20 days**	**Wed 10/3/18**	**Tue 10/30/18**	**100%**
13	2.1.2.1 In-house Pull Test Setup	20 days	Wed 10/3/18	Tue 10/30/18	100%
14	2.1.2.2 In-house preparation of polyurethane coated samples	20 days	Wed 10/3/18	Tue 10/30/18	100%
15	2.2 Testing bond strength of after IR treatment	30 days	Wed 12/26/18	Tue 2/5/19	100%
16	**2.3 Testing bond strength after ultrasonication**	**160 days**	**Wed 8/22/18**	**Tue 4/2/19**	**100%**
17	2.3.1 Establishment of industry partner for Ultrasonic testings	30 days	Wed 8/22/18	Tue 10/2/18	100%
18	2.3.2 Sending samples to carryout ultrasound tests	30 days	Wed 10/3/18	Tue 11/13/18	100%
19	2.3.3 Getting quatations and finalizing a Ultrasonic machine	10 days	Wed 11/14/18	Tue 11/27/18	100%
20	2.3.4 Setting up the machine and getting the proper training	30 days	Wed 11/28/18	Tue 1/8/19	100%
21	2.3.5 Testing the samples(AFRL and In-house coated samples)	60 days	Wed 1/9/19	Tue 4/2/19	100%
22	2.4 Testing bond strength after IR-Sonication treatment	86 days	Wed 4/3/19	Wed 7/31/19	100%
23	2.5 Monitoring temperature profile across the underlying substrate	90 days	Wed 10/31/18	Tue 3/5/19	100%
24	2.6 Infrared thermography for spot temperature measurements	30 days	Wed 3/6/19	Tue 4/16/19	100%
25	2.7 M 2.1 50-25% reductionin the peel-strength	0 days	Wed 4/3/19	Wed 4/3/19	100%
26	**3 Fabrication and Testing of IR-Sonication system**	**120 days**	**Wed 1/9/19**	**Tue 6/25/19**	**100%**
27	**3.1 Design and Fabrication of IR-Sonication probe**	**120 days**	**Wed 1/9/19**	**Tue 6/25/19**	**100%**
28	3.1.1 3D Design of IR device	30 days	Wed 4/3/19	Tue 5/14/19	100%
29	3.1.2 3D Design of Sonication Device	30 days	Wed 1/9/19	Tue 2/19/19	100%
30	3.1.3 3D Design of the Integrated system	60 days	Wed 4/3/19	Tue 6/25/19	100%
31	**4 Final Design and Economic Analysis**	**206 days**	**Thu 11/8/18**	**Thu 8/22/19**	**100%**
32	4.1 Characterization of Substrate after treatment	1 day	Wed 4/3/19	Wed 4/3/19	100%
33	4.2 Quarterly Progress Report - 1	10 days	Thu 11/8/18	Wed 11/21/18	100%
34	4.3 Quarterly Progress Report - 2	10 days	Fri 2/8/19	Thu 2/21/19	100%
35	4.4 Quarterly Progress Report - 3	10 days	Fri 5/10/19	Thu 5/23/19	100%
36	4.5 Costing and Economic Analysis	30 days	Fri 5/24/19	Thu 7/4/19	100%
37	4.6 Final technical report	10 days	Fri 8/9/19	Thu 8/22/19	100%



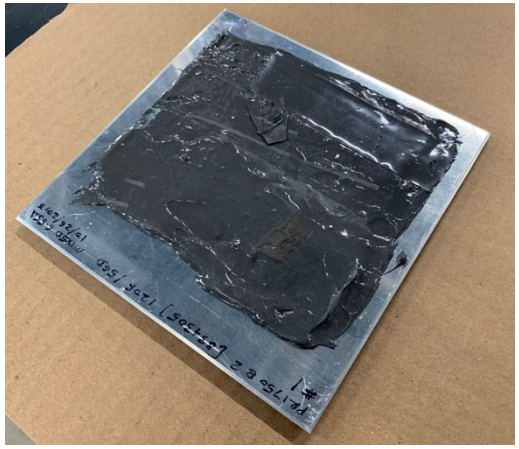
Tested sample types

1) **AFRL-**RE coating on aluminum substrate: Coating thickness, 0.5 mm
2) **AFRL-**RE coating on aluminum substrate: Coating thickness, 1 mm
3) **AFRL-**RE coating on composite substrate: Coating thickness, 0.5 mm
4) **AFRL-**RE coating on composite substrate: Coating thickness, 1 mm
5) **AFRL-**FP coating on aluminum substrate: Coating thickness, 0.5 mm
6) **AFRL-**FP coating on aluminum substrate: Coating thickness, 1 mm
7) **AFRL-**FP coating on composite substrate: Coating thickness, 0.5 mm
8) **AFRL-**FP coating on composite substrate: Coating thickness, 1 mm
9) **NAVAIR**-sealant coated samples: Coating thickness, 1-2 mm
10) **Lab coated** samples on aluminum substrate: Spray coated (1 mm)
11) **Lab coated** samples on aluminum substrate: Dip coated (1-2 mm)

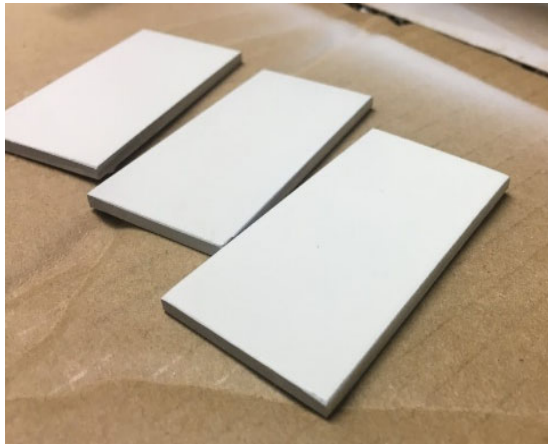
Tested Samples



AFRL coated sample with DOD specialty coating,

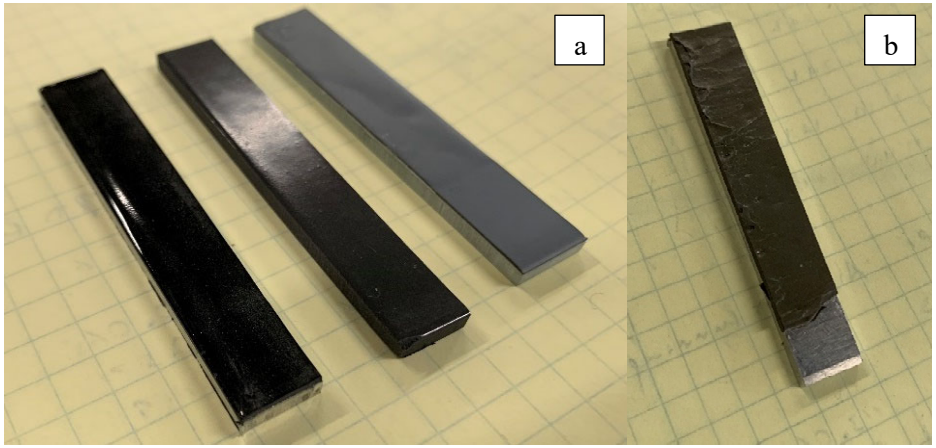


NAVAIR coated sealant sample



In-house spray painted samples

- 3”x 0.5” coupons were chosen considering the peel tester machine capabilities

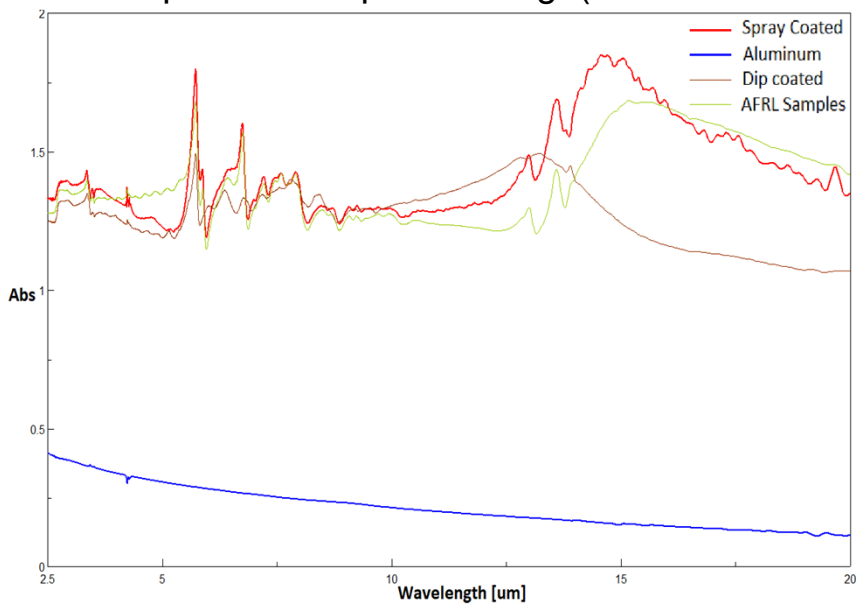


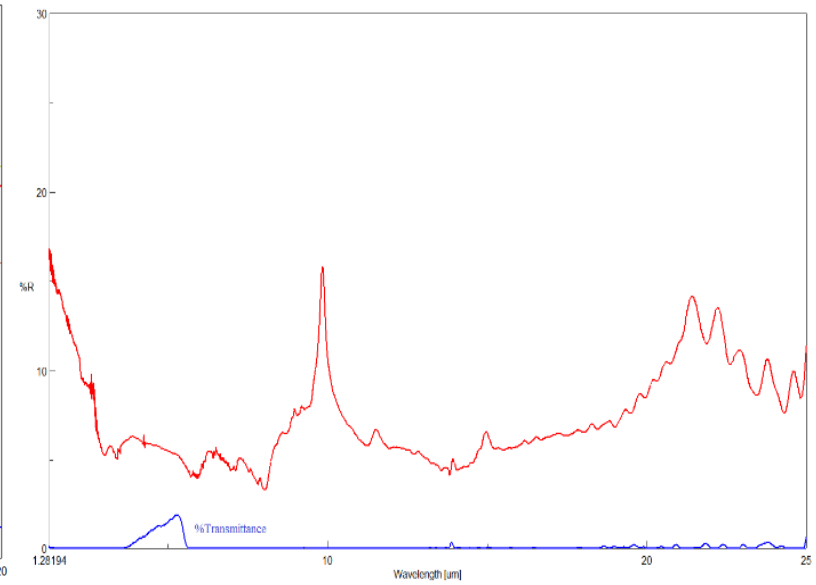
Rectangular coupons of 3”x 0.5” dimension were made from a) AFRL and NAVAIR

Results(Task 1)

Characterization of the surface coat and the underlying substrate coat

- Optimum IR spectral range(Both MW-IR and SW-IR regions)

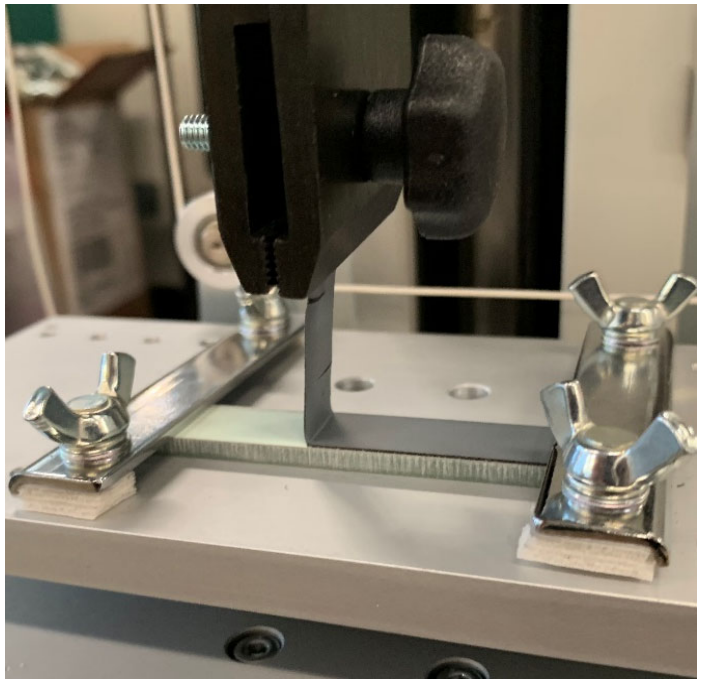


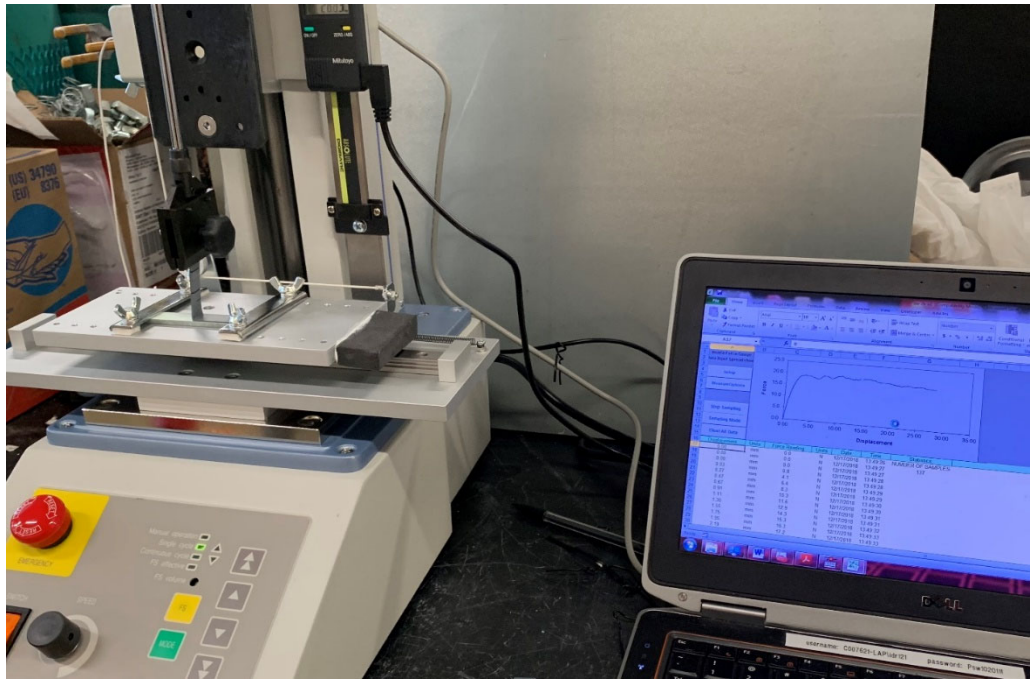


% Absorption of IR spectra of coated and uncoated substrates, %Reflectance and %Transmission of the rain erosion coating material

- Complete attenuation of IR radiation in the SW and a part of MW, however it shows some transmittance in the 3.4 μm to 5.7 μm range.
- SW: 0.7 – 2.5
- MW: 2.5 - 5

Results(Task 2)





Peel tester machine (ASTM D429 compatible test)

- Peel tester was used to determine the bonding strength of the as-received and treated samples

Untreated Samples

Sample Name	Average Peel* Force As-received (N)
AFRL-RE coating on aluminum substrate: Coating thickness, 0.5 mm	28 N
AFRL-RE coating on aluminum substrate: Coating thickness, 1 mm	58 N
AFRL-RE coating on composite substrate: Coating thickness, 0.5 mm	40 N
AFRL-FP coating on aluminum substrate: Coating thickness, 1 mm	70 N

- **FP samples have a higher binding strength than the corresponding RE samples**
- **As-received composite samples have a higher binding strength than the corresponding aluminum samples**

** Experiments were done in triplicate, percentage error* ± 5%
*** FP-composite 0.5 & 1 mm as-received samples and FP-aluminum 0.5 mm as-received samples showed cohesive failure*

IR Experiments

IR emitter unit with SW-IR and MW-IR modules at Radiant Energy Systems Facility

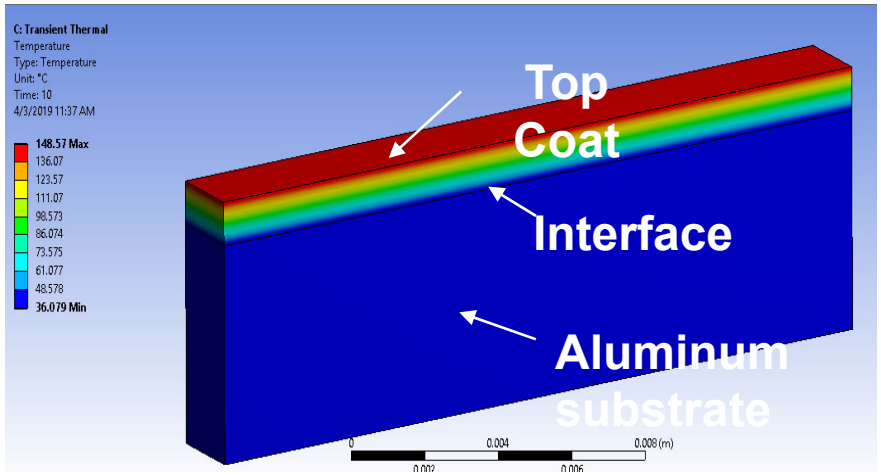
- Experiments were carried out at the R&D facility in Florida as well as at the Radiant Energy Systems (Industrial Partner) facility.

IR emitter unit with interchangeable SW-IR and MW-IR cassettes (in-house)

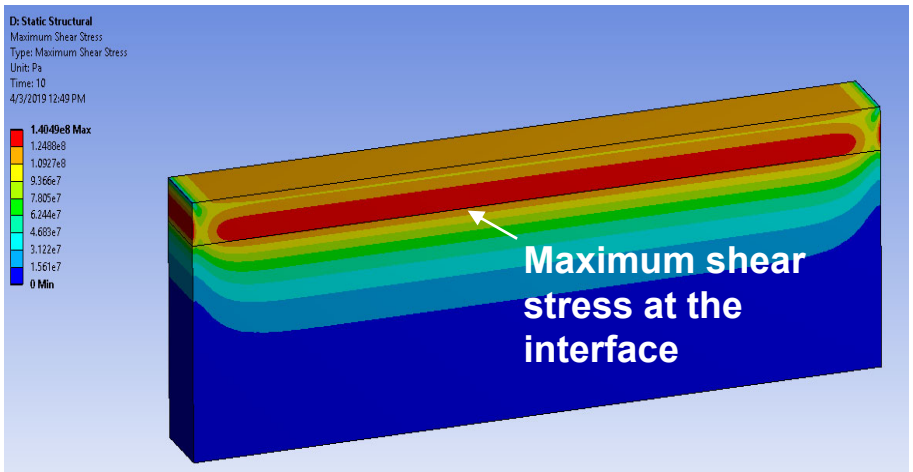
Computational Simulation

OBJECTIVES:

1) To determine temperature profile across the substrate with varying IR intensity
2) Determine shear stress at the interface
3) Validation of computational modeling with experimental data

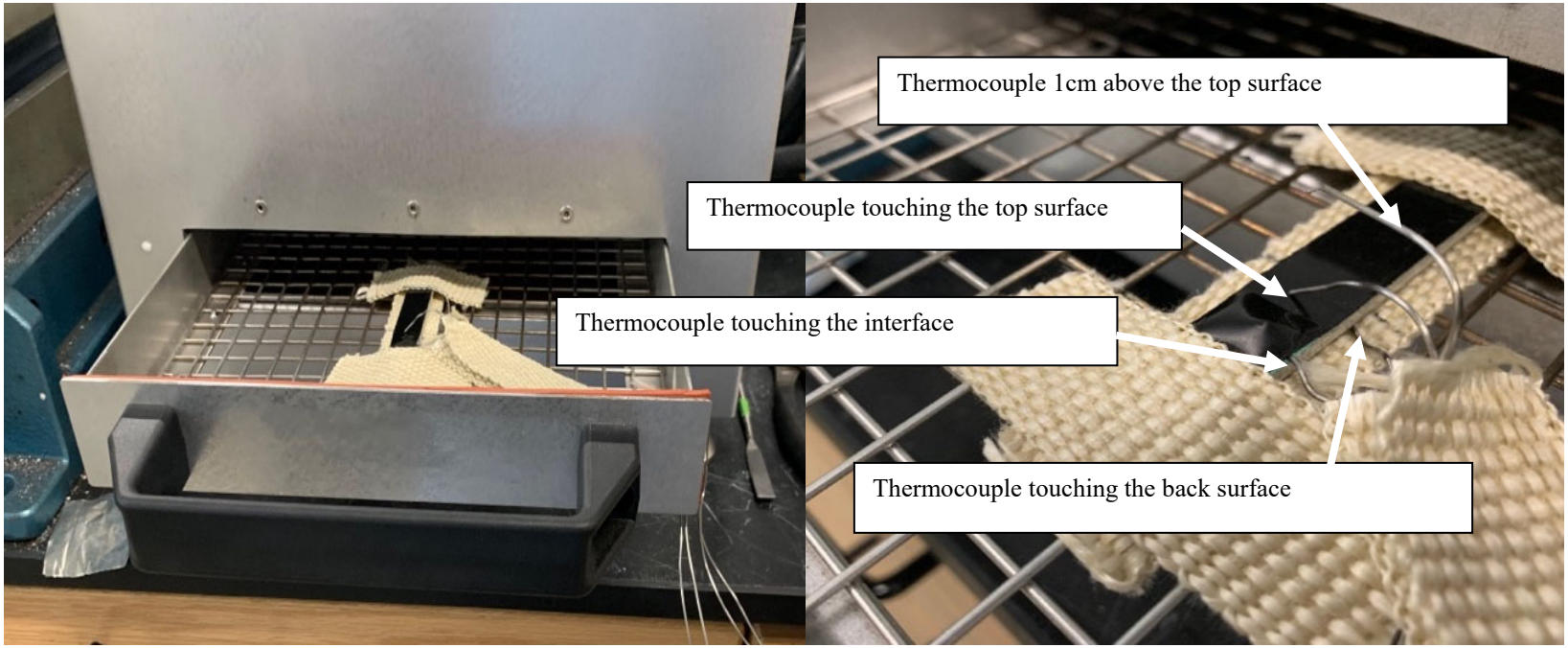


- Transient model was created in ANSYS.
- Transient temperature data (75% intensity) was used for the structural model to generate the shear stresses.
- Transient Model was done for a 10s, 12S, 20S period with varying IR intensity and with varying top coat thickness.
- The maximum shear stress was seen in the interfacial layer.



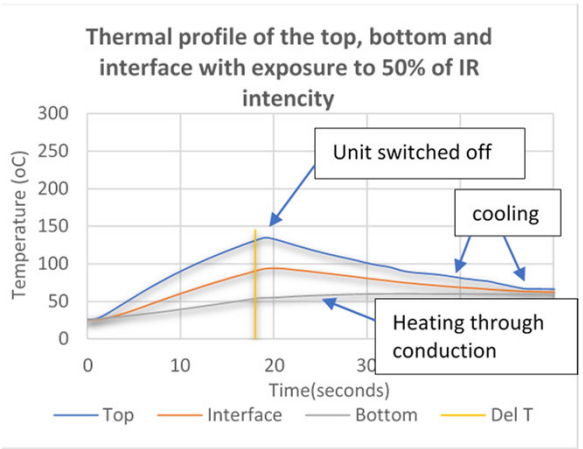
Temperature Profile of the samples under IR irradiance

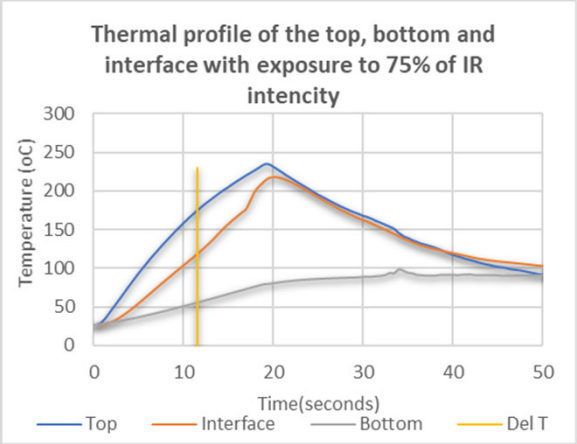
Objective:
1) Computational result validation
2) Determine optimum IR exposure time

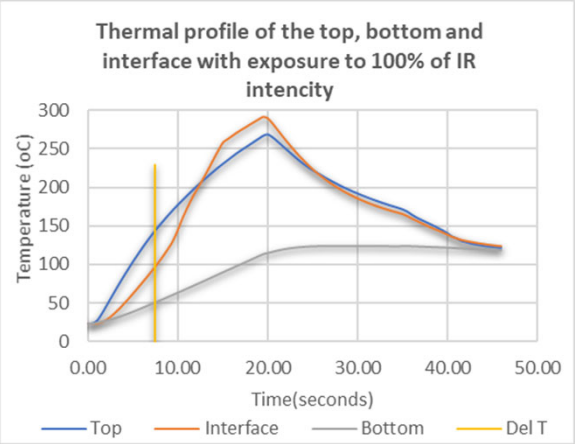


Thermocouple arrangement for temperature profile monitoring

Temperature Profiles





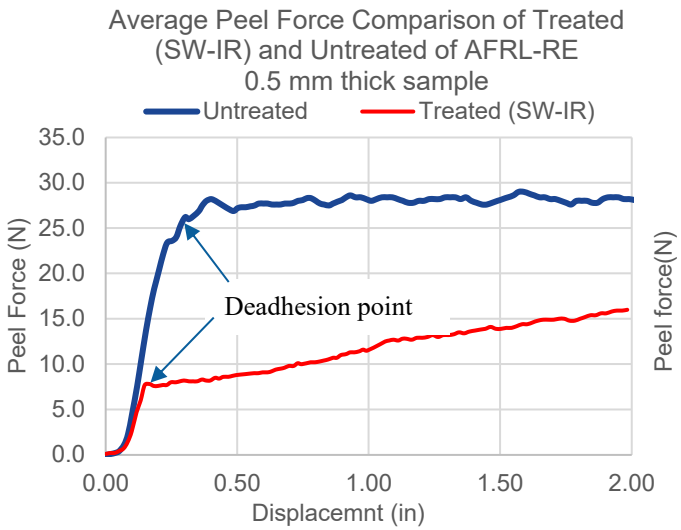


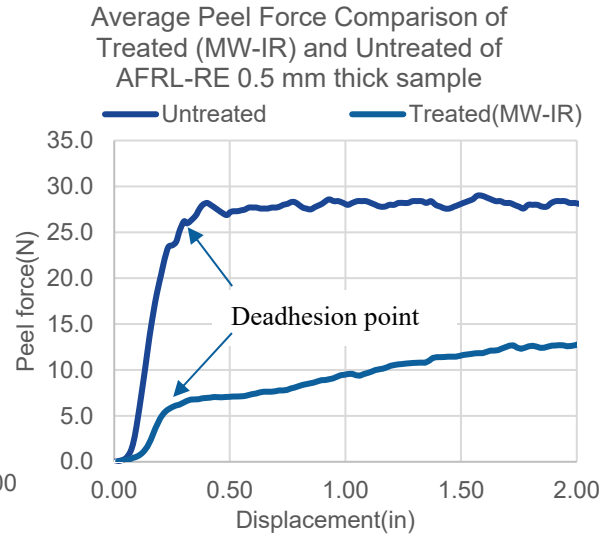
- 100% IR intensity ~ 4800W
- Thermocouple readings of the top, bottom and interface were recorded using LabVIew.
- IR heating was applied for 20 seconds with varying intensity.
- Temperature difference between top surface and interface were in the range of 45-55°C
- Interface temperature were in the range of 90°C-110°C
- Time to reach the maximum temperature difference were 18s , 12s and 8s for 50% intensity, 75% intensity and 100% intensity respectively (indicated in dotted line Del T).
- Exposure time was selected based on these experiments

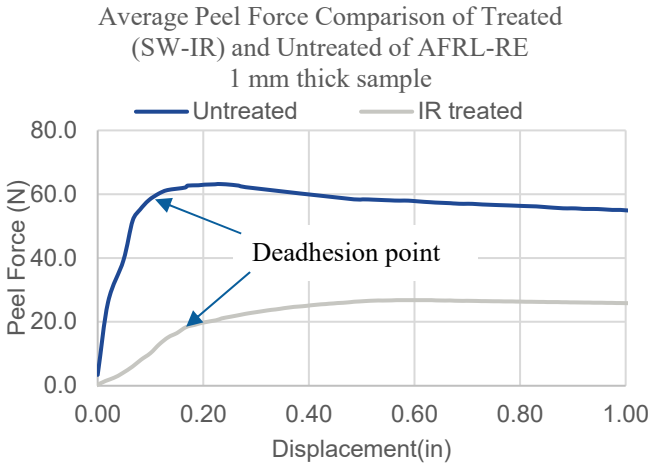
MW-IR and SW-IR experiments

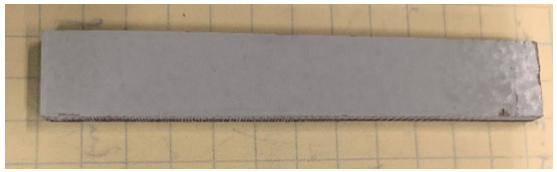
RE Coatings on Aluminum substrates

- RE 0.5 mm thick coated samples were tested
- Peel reduction by **76%** was achieved with SW or 3.6 times lower peel strength with treated samples
- Peel reduction by **79%** was achieved with MW or 4.7 times lower peel strength with treated samples
- As-received 1mm Al-RE samples showed twice the bonding strength (62N compared to 28N) than 0.5 mm thick substrate.
- Even then, the peel strength reduction after SW-IR exposure is more than 3 times when compared to the as-received samples





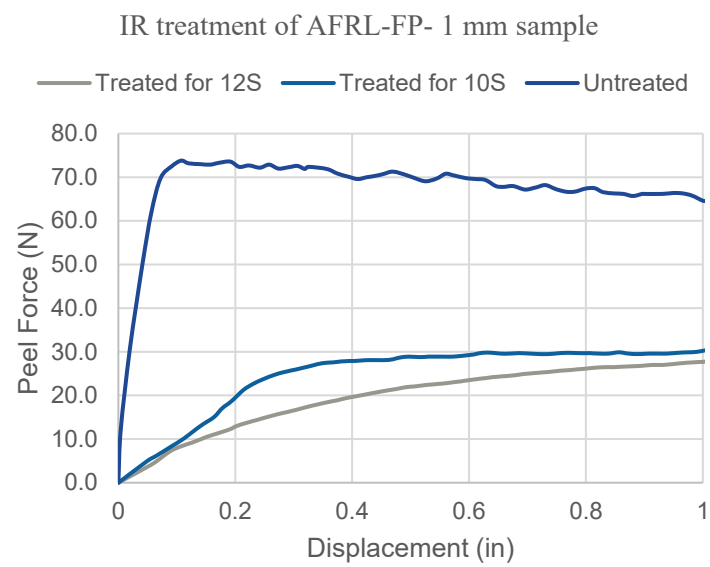




Peeled samples

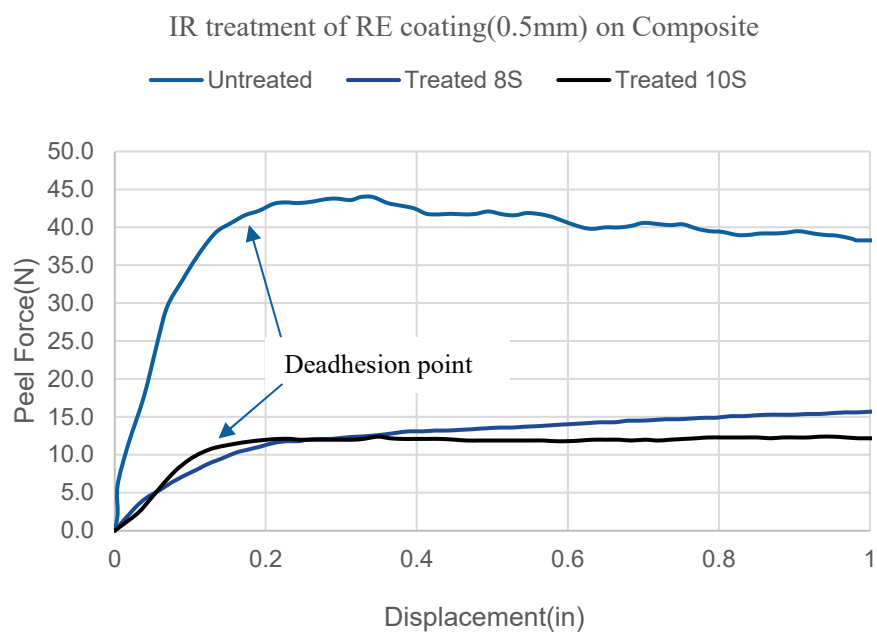
AFRL-FP Coatings

- We also tested the peel of as-received AFRL- FP coated samples (1mm thickness).
- We faced difficulty in getting the base case(without treatments) results as these samples showed higher bonding strength between the coating and the underlying substrate than compared to the RE coatings
- On exposing these samples to SW-IR for 10s and 12s, the peel strength reduction was found to decrease by more than **3.5 times**.

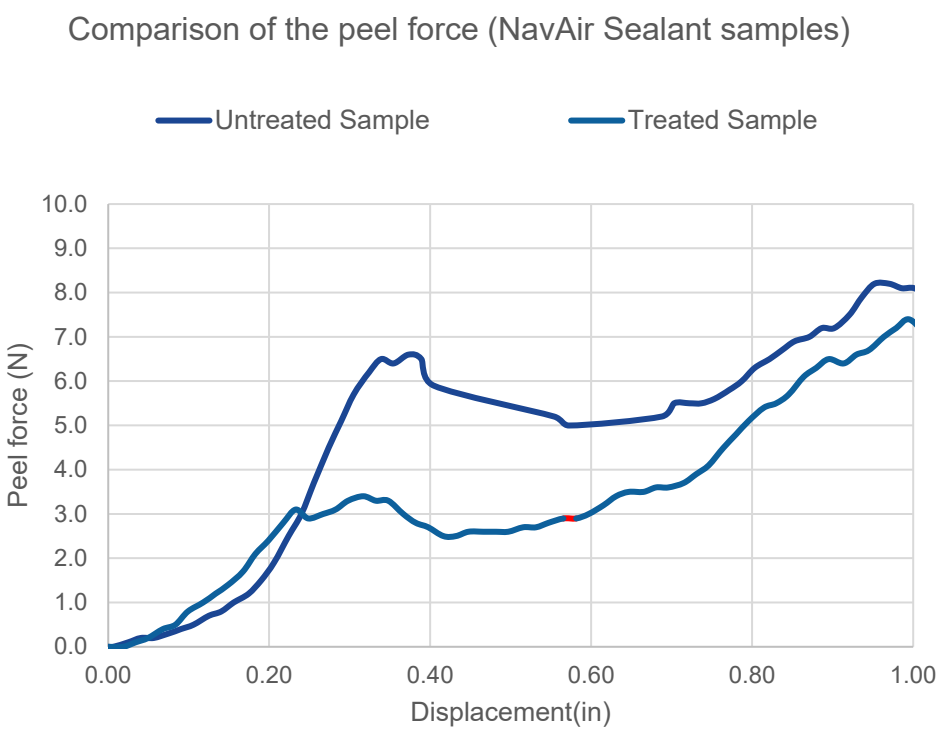


RE Coating on composite substrate

- Due to the abrasiveness of the composite material, it was very difficult to cut and machine these samples
- Average peel strength of the AFRL-RE composite substrate, of 0.5mm coating thickness, was greater than the average peel strength of the same coating of Aluminum substrate.
- On exposure to SW-IR, the reduction in the peel strength was greater than 3 times, i.e., in the range of 60-70%.
- Both IR exposure of 8 S and 10 S gave similar results.



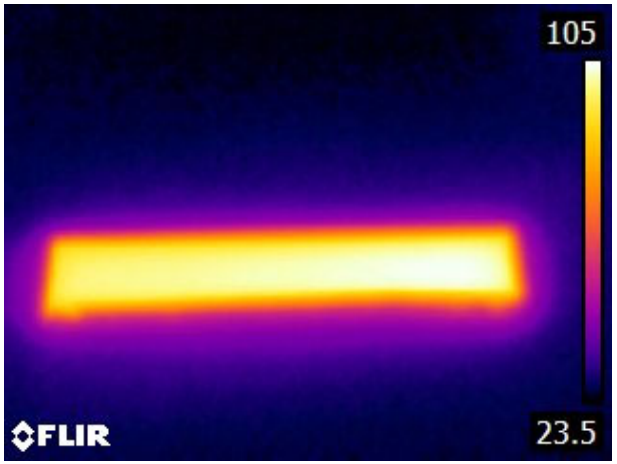
Sealant Sample provided by NavAir



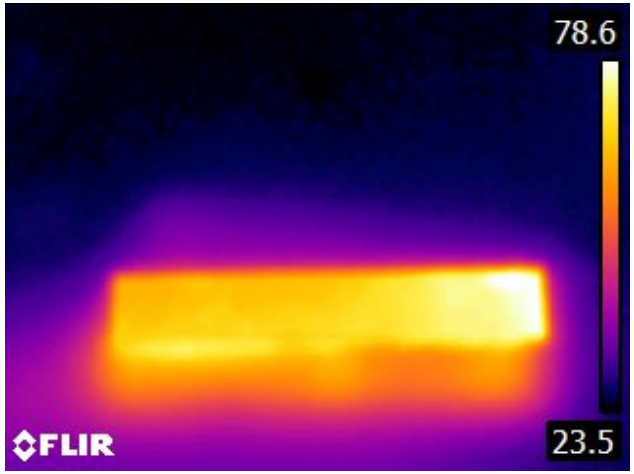
Peeled samples

- The sealant samples from NAVAIR have irregular thickness, the peel strength reduction of all the Nav-air samples were in the range of 50%.

Thermal Imaging

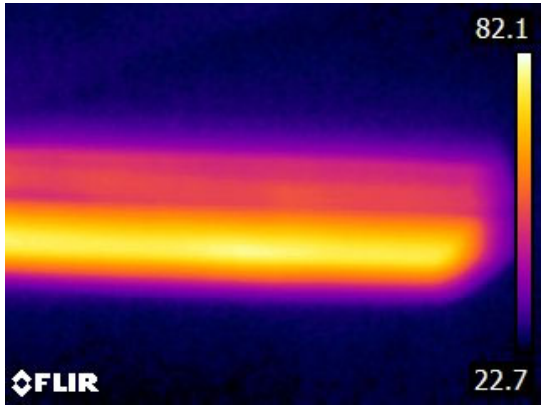


Top Surface (shows a reading of maximum105°C)



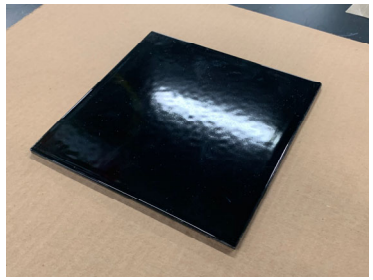
Bottom Surface (shows a reading of maximum 78.6°C)

- These images were taken approximately 5s after taking out from the IR furnace.
- There was a clear gap in temperature on the top coat, where it was exposed to IR directly, and the bottom substrate surface.

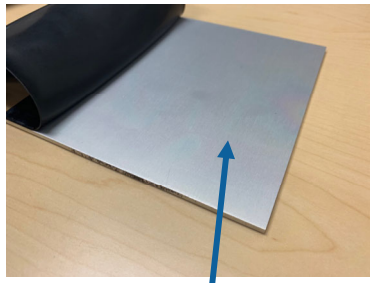


Cross section

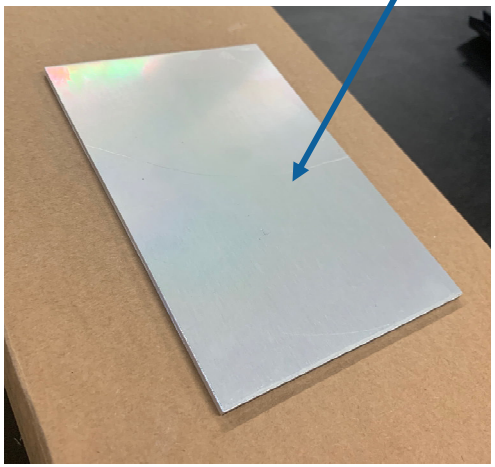
Peeled samples: Video & photos



6”x6” sample



Peeled sample after IR treatment



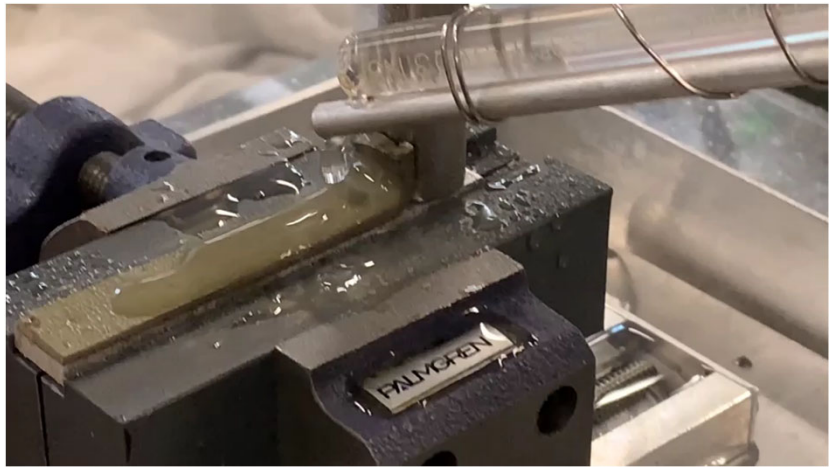
- Objective: Testing the theory with a larger plate
- Sample: AFRL-RE 6”x6” of 1 mm thick plate
- 1 mm thick AFRL-RE coating on aluminum plate (6”x6”)
- IR Intensity 100% (4800W) for 12 seconds
- Pliers were used to peel the top coat

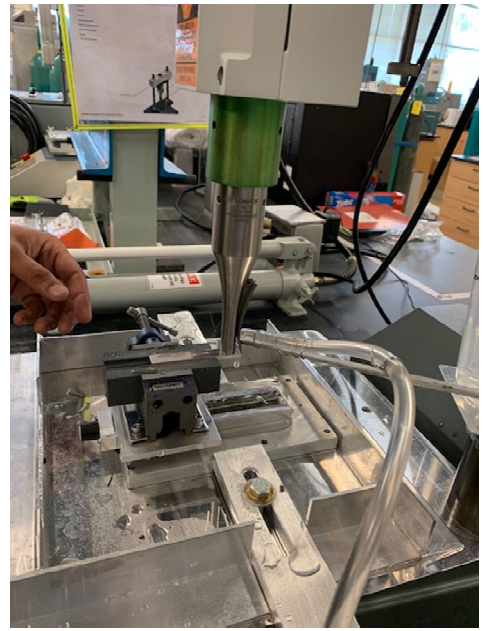
Ultrasonic Experiments

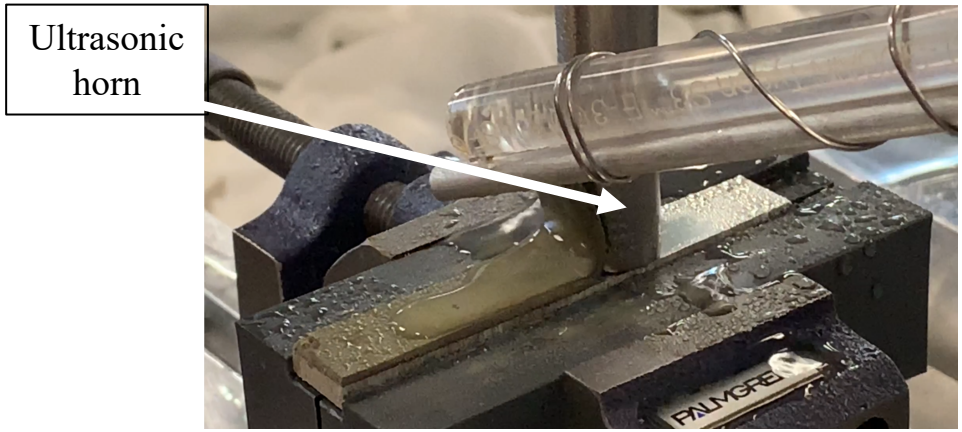
- Two types of experiments were carried out
  - Directly applying ultrasonic energy to the Top layer of the sample (Friction stir heating)
  - Maintaining a small gap between the horn and the top layer with a film of water or ultrasonic gel (through cavitation)

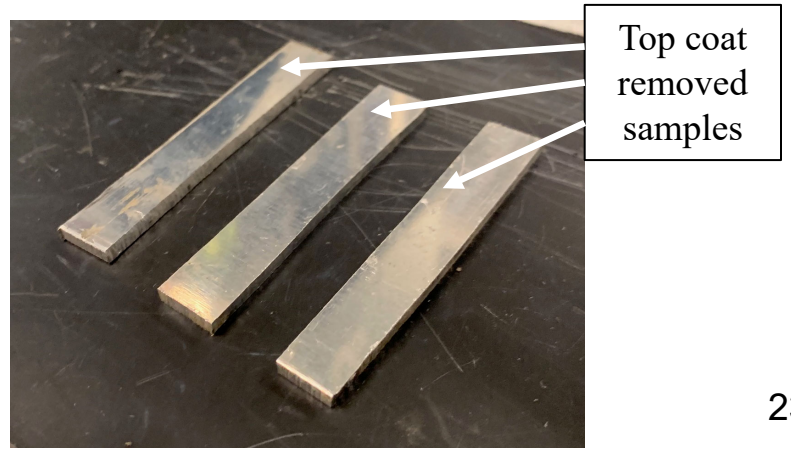
Ultrasonic experiments: Video & photos

Video of coating removal by ultrasonication







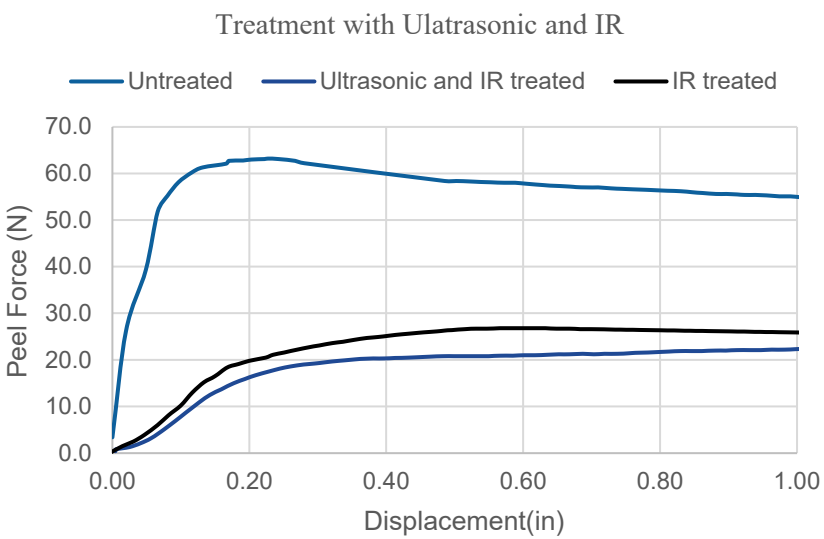


Results (Task 3)

IR-Sonication synergy: Hybrid approach

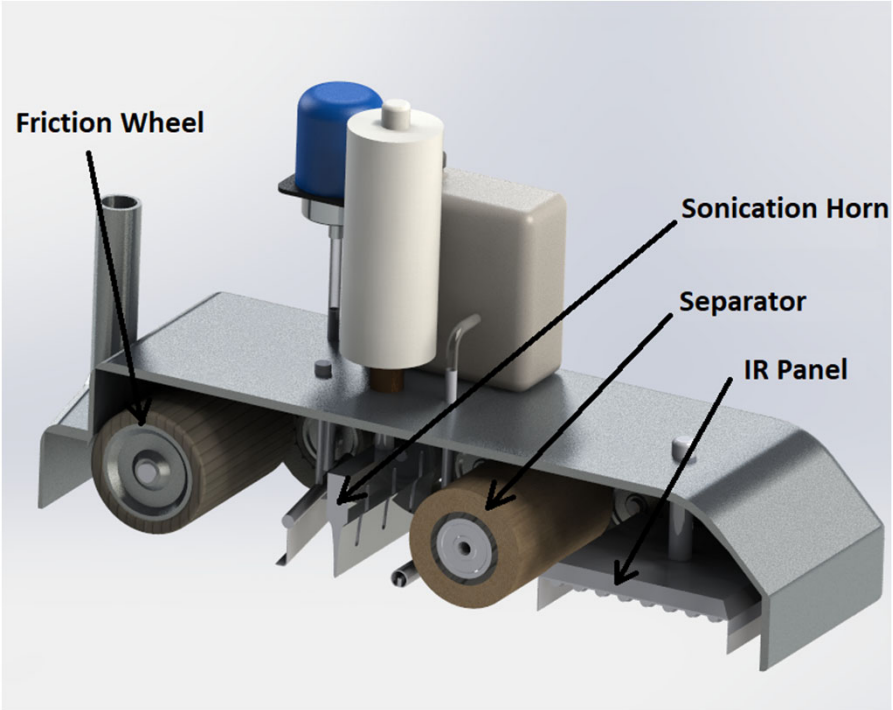
Thickness	Untreated (N)	IR treated (N)	Ultrasonic Treated (N)	Hybrid (N)
AFRL-RE - 20 mil	35-45	12-15	30 -35	11-13
AFRL -RE- 40 mil	60-65	20-22	45-40	18-20

- Additional Percentage reduction of 5-10% was observed with the hybrid approach compared to the best-case IR alone approach.
- We have tried several scenarios with different IR, sonication sequence. Introducing ultrasonication first and then IR gave us better results than exposing to IR first and then ultrasonication.



IR-sonication exposure effect on peel strength of AFRL-RE sample of 1 mm thickness

Modelled IR –sonication device



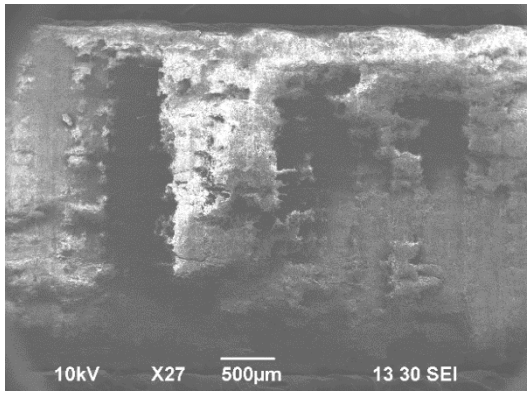
SolidWorks Model of IR-sonication device

- A portable unit as well as a unit operated by a robotic arm were modelled in SolidWorks.
- This unit capable of providing IR or ultrasonic independently or simultaneously.
- IR panel can be made in an easily interchangeable manner to switch between SW-IR heater cassette and MW-IR heater cassette depending on the requirement.

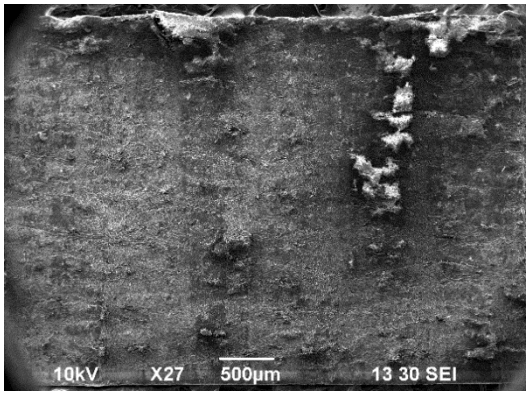
Results(Task 4)

Structural Integrity of the underlying substrates: SEM analysis

- No visible structural damage was noticed in the SEM images for composite substrate
- No cracks or delamination was observed on the surface and in the cross-section of the underlying substrates.
- These images clearly indicated that no damage has been induced in the underlying substrates from the IR treatment

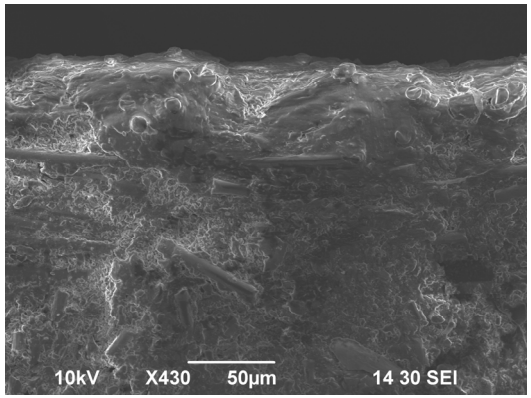


Untreated

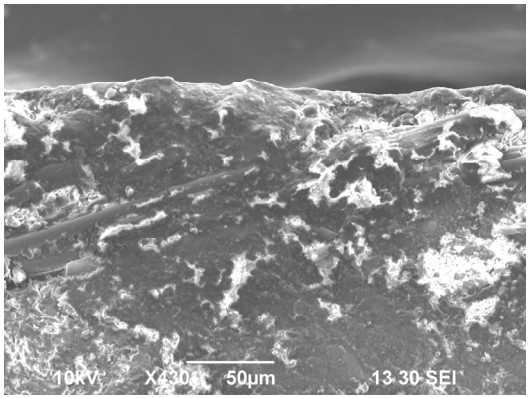


Treated

Cross section



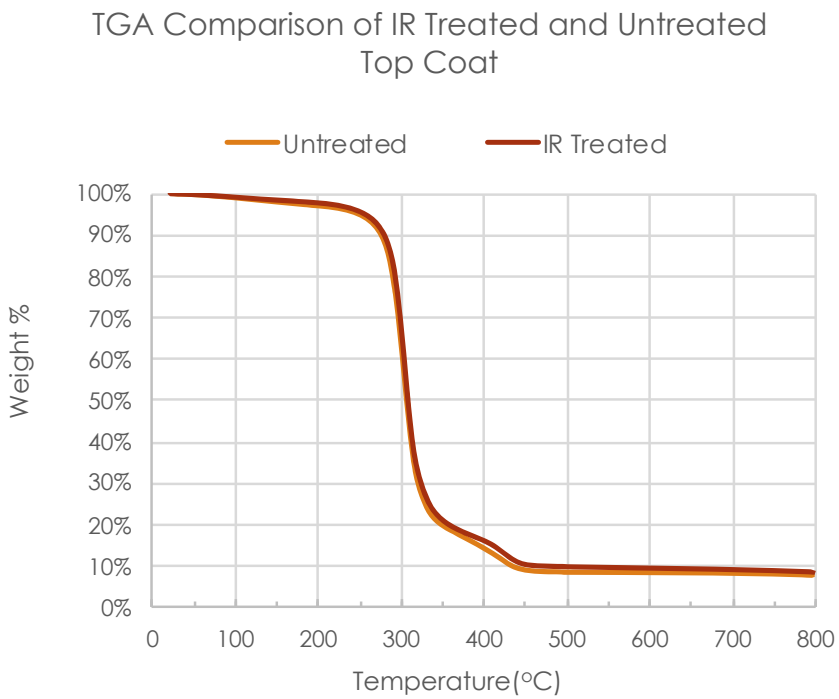
Untreated

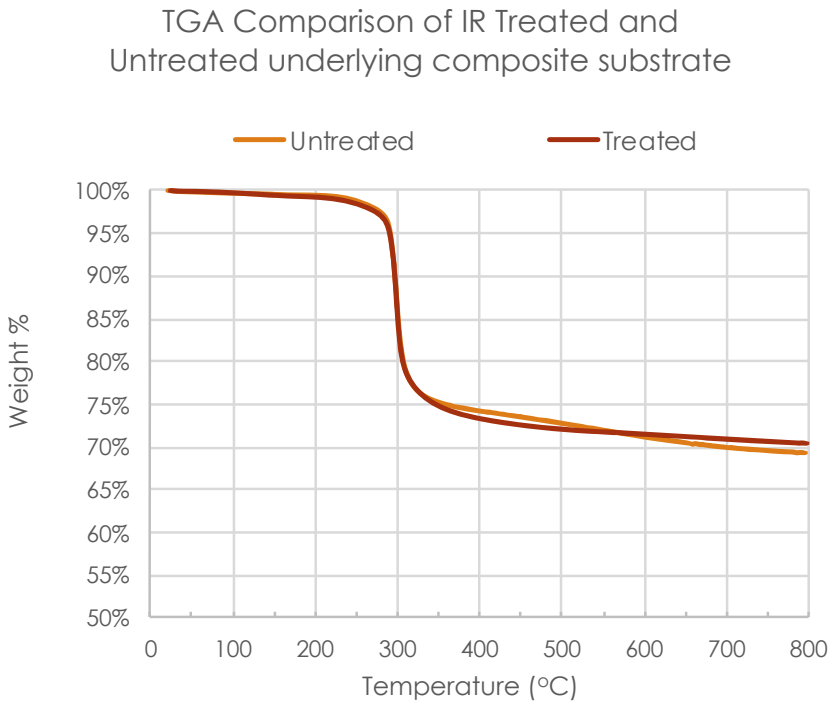


Treated

Top surface

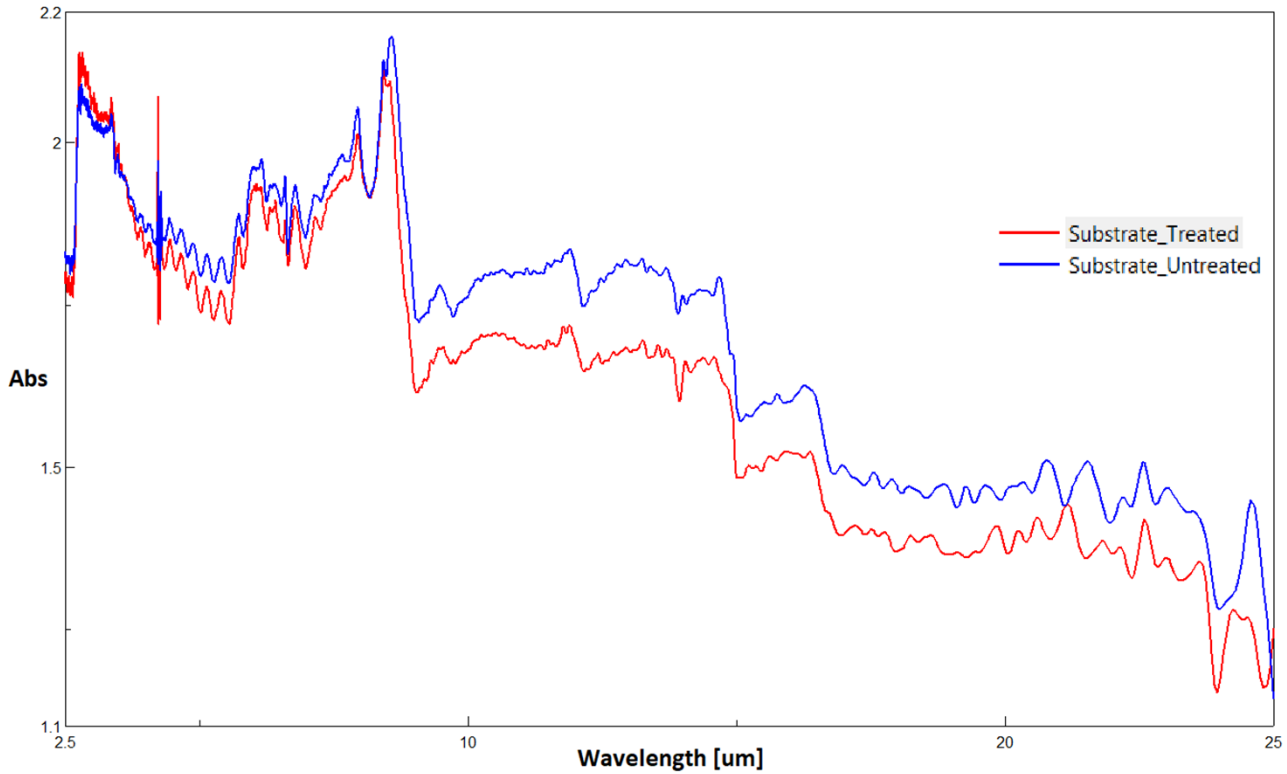
Structural integrity: TGA comparison of Treated and untreated samples





- IR treated and untreated samples have shown no difference.
- Not only the substrate, even the top coat, which is directly exposed to IR radiation, has shown no impact at all.
- The main reason is the operating temperatures of these samples which were exposed to IR were significantly lower than their degradation temperatures.

Structural integrity: IR comparison of Treated and untreated samples



- FTIR analysis shows that IR treated and untreated samples have the same absorption peaks throughout the range.
- This is an indication that the surface properties of the substrate remained unchanged due to the IR exposure.

Next Steps

- Project objectives were successfully achieved.
- 75-50% Interfacial bond reduction was achieved using the IR/Ultrasonication treatments.

Technology Transfer

- A Poster on “Introducing Interfacial Bond Failure in Thick Elastomer Coatings by Tailored Radiation Approach” will be presented in the 2019 SERDP & ESTCP Symposium(12/3/2019).

- Two manuscripts will be submitted.

Key Points

- The use of IR is a very effective noncontact approach to remove coatings without impacting the underlying substrate.
- Sonication augments interfacial bond failure introduced by IR.
- No waste generation
- Due to the quick response time and the ability to finetune according to the absorption of the top surface makes this a very unique and effective approach when it comes to reducing the adhesion between any layers.
- This technique can be adapted to remove stickers, plastic lamination and any kind top layer removal applications.
- More importantly this approach will make sure to keep the underlying substrate in its original condition while being ergonomically less challenging.

Future Research

- Since the expected results through IR/Sonication treatments exceeded the set project milestones, it's essential to design and develop a semi or fully automated unit to remove coatings from larger and curved surfaces with different thicknesses.

- We have identified the optimum IR exposure levels with different coating thicknesses. So our next approach should be to incorporated this to our removing mechanism.

- An automated process should be developed to vary the IR intensity(or exposure time) according to the feedback from the thickness sensor placed before the IR heaters.

Future Research

- Develop a small curved ultrasonic horn with the assistance of an Industrial partner*, who has expertise on developing ultrasonic horns, capable of removing corners and bends (Specially considering NAVAIR requirements)

- Develop a semi/fully automated mechanism to cater flat, concave and convex samples and complex structures/geometries (i.e. fasteners)

**Sonics & Materials, Inc is an industrial partner who has shown keen interest in working with us*

BACKUP SLIDES

Publications

- First Manuscript will be submitted before the end of December 2019

Project Funding (single year)

Only for Molekule

	2018/2019
Funds received to date ($K)	$128,534.00
% Expended	$126,867.04
Funds Remaining ($K)	$1,666.96

Status of Funds for Federal Performers

- AFRL is the Federal Performer for the Project

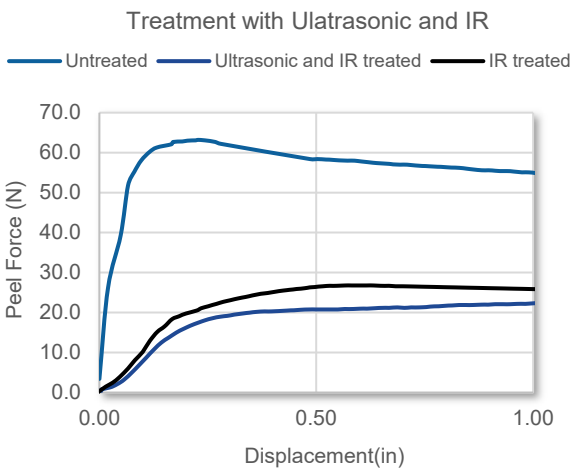
FY2018/2019 Funds			
Directly Funded Federal Performer(s)	**Funds Received**	**Funds Obligated***	**Percent Funding Obligated**
AFRL	12,000	12,000	100%

* Funds put on contracts and/or purchase orders that have been issued, and funds associated with internal labor or travel expenses that have been incurred.

WP-1347: INTRODUCING INTERFACIAL BOND FAILURE AT THE ELASTOMER-PRIMER INTERFACE USING TAILORED INFRARED-SONICATION HYBRID TECHNOLOGY

Performers: Jaspreet Dhau & Chatura Wikramaratne

Molekule Inc.

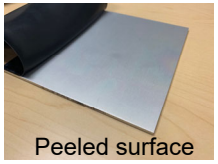


Technology Focus

- *A technology that can effectively removes thick elastomeric coatings and treatments without damaging the underlying substrate or causing environmental problems.*

Research Objectives

- *Develop an innovative and cost-effective ergonomic Infrared-Sonication hybrid technology for removing thick, elastomeric specialty coatings used on DOD Weapon Systems.*


Peeled surface

Project Progress and Results

- *All the milestones were achieved.*
- *One of the key significant millstone of 50-25% reduction in the peel-strength of the elastomeric coatings after IR-Sonication treatment (Milestone 2.1) was achieved with far better results than we initially proposed. The average peel strength reduction for all the coatings types and thickness tested were in the **75-50%** range.*

Technology Transition

- *A Poster will be presented in the 2019 SERDP & ESTCP Symposium(12/3/2019).*
- *First manuscript will be ready for submission before the end of December 2019*