This presentation does not contain any proprietary, confidential, or otherwise restricted information

Active, Tailorable Adhesives for Dissimilar Material Bonding, Repair and Assembly

Dr. Mahmoodul Haq[1] and Dr. Lawrence T Drzal[2]

[1] Assistant Professor, Civil & Environmental Engineering

[2] University Distinguished Professor, Chemical Engg. & Material Science

Composite Vehicle Research Center (CVRC)

Michigan State University

Project ID # : LM087

MICHIGAN STATE UNIVERSITY | Composite Vehicle Research Center

OVERVIEW

TIMELINE

- **Start Date:** October 1, 2013
- **End Date:** September 30, 2016
- Percent Complete – 75%

BUDGET

- **Total Project Funding:** $599,999
- **Funding Received in Budget Period 3: (01/2015 – 12/2015) :** $236,629
- **Funding for Budget Period 4: (01/2016 – 09/2016) :** $0

BARRIERS ADDRESSED

- **Joining and Assembly**
  - **Light-weight, reversible bonded joints**
- **Performance**
  - **Enhanced Damage Resistance of Joints using nanoparticles**
- **Predictive Modeling Tools**
  - **Development of Experimentally Validated Simulations.**

Partners / Collaborations

- **Eaton Innovation Center, MI.**

Project Lead

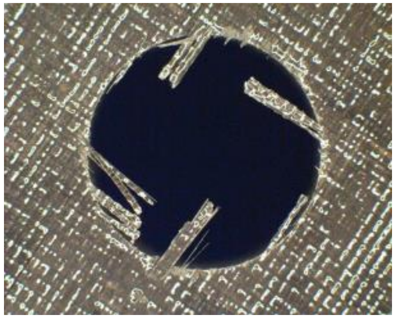
- **Michigan State University, Composite Vehicle Research Center (CVRC).**

Introduction / Relevance - Joining

- **JOINING / ASSEMBLY**

- Joining is inevitable, allows versatility in assembly and repair, reduces costs and time.
- Considered a 'weak-link' in the structure due to complex phenomena & interactions.

- ***Mechanical Fastening***
- ***PROS: a) Repair and Re-assembly, b) confidence in use as it is commonly used***
- ***CONS : a) Adds Weight, b) machining holes, c) delamination in composites, d) stress-concentrations***



Delamination in composites due to hole-drilling, Gardiner, Composites World , (2012)

- ***Adhesive Bonding***
- ***PROS: a) Light Weight and b) load distribution over larger areas***
- ***CONS : a) permanent joint (cannot be repaired or re-assembled), b) lack of confidence in common use to reliability of bonding.***

Examples of Adhesive Joints
a) Lap-Joint , b) Double Lap-Joint

There is a Need for a JOINING TECHNIQUE that can INHERIT the MERITS of BOTH bolted & bonded techniques while still being compatible with current assembly line practices

Introduction / Relevance - Joining

RELEVANCE:

Key Technical Gaps for Systems for Light-Duty Vehicles

System	Three Most Significant Technical Gaps Impeding Widespread Implementation		
Body Structures (Composites)	Lack of understanding of properties with respect to fracture and energy absorption	Lack of predictive engineering and modeling tools	Lack of high-volume manufacturing capability
Body Structures (Metals)	Lack of technology for joining dissimilar materials	Properties of alternative lightweighting materials are inadequate for forming and energy absorption	Modeling, simulation, and design tools are inadequate for optimization
Chassis and Suspension	Inadequate properties (strength, ductility, corrosion resistance, etc.)	Manufacturing capacity to produce high- integrity components is inadequate	Robust joining processes, especially to other materials, are lacking
Closures, Fenders, and Bumpers	Fast and reliable processes for joining dissimilar materials are not available	Design knowledge and databases are inadequate	Cost/availability of most lightweight materials and current manufacturing processes are not competitive
Engines and Transmissions	Materials needed for advanced technology propulsion systems are not cost competitive	Properties of current materials are not adequate	Databases for modeling and design are inadequate

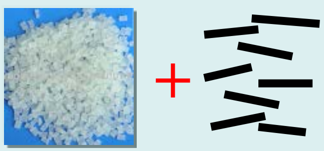
Source: http://www1.eere.energy.gov/vehiclesandfuels/pdfs/wr_ldvehicles.pdf

This project address three concerns on : a) joining dissimilar materials, b) experimentally validated simulations and c) joining techniques relevant and capable of easy transition to industrial applications

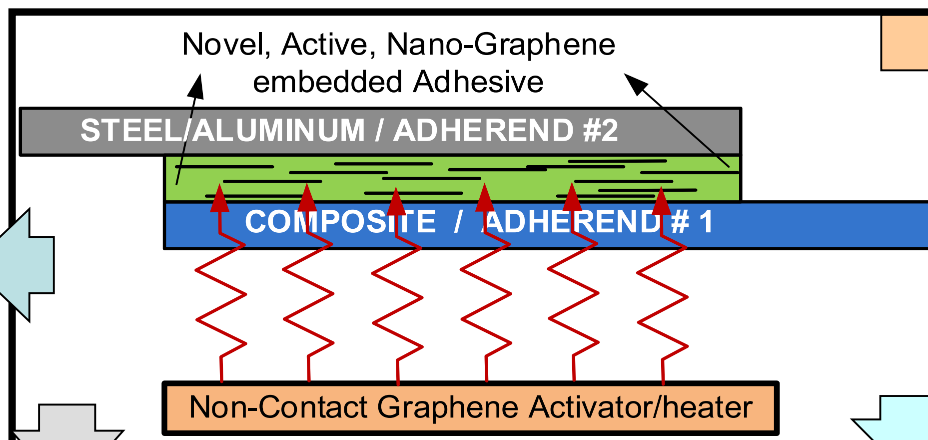
Summary of Progress : Objective, Approach, Relevance, Milestones and Accomplishments

- ***OBJECTIVE:*** *To demonstrate the feasibility of 'ACTIVE Adhesive' technology for structural joining of similar / dissimilar substrate materials.*

A. Processing, Material Development & Optimization

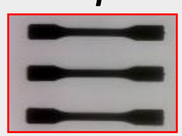


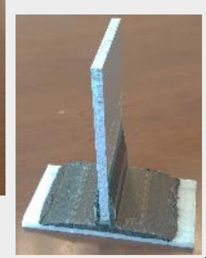
Thermoplastics + GnP =
Active Adhesive Pellets and Films



E. Dissemination of Results
Journals +
Conferences +
Invited talks +
Roadmaps

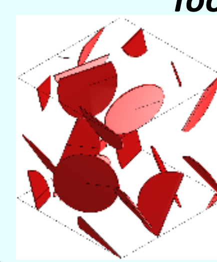
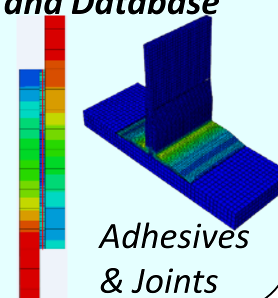
B. Lab-scale Evaluation & Experimental Characterization





Adhesives & Joints

C. Development of Design Tools and Database

Adhesives & Joints

- ***MATERIALS USED:***

Substrates:	**TP Adhesives:**
➢ Aluminum	➢ Nylon-6
➢ Steel	➢ Polycarbonate
➢ CFRP	➢ Polyolefins
➢ GFRP	➢ ABS (current)

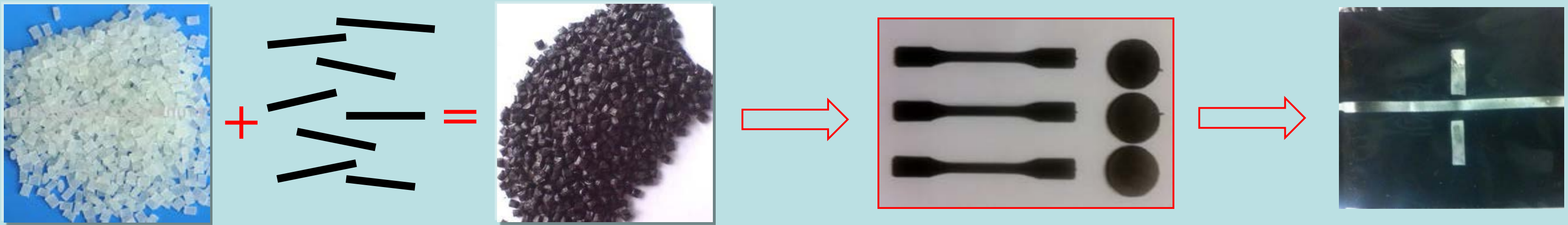
- **APPROACH:** An integrated experimental and numerical computational materials (materials by design) based approach. Multi-use, Repair & Reassembly?

Summary of Progress : Relevance, Milestones and Accomplishments

	Milestone	Type	Description	Status
FY 14	Activation and Bonding	Technical	The novel active adhesive couples with microwave radiations to activate, bond/un-bond resulting similar joints	SUCCESS!!
FY 14	Structural Properties Defined	Go / No-Go	The novel active adhesive structural properties (lap-shear) pre- and post- exposure to corrosive environments is better or equal to requirements in industrial practices with conventional bonding techniques	SUCCESS! GO ☑
FY 15	Demonstration of Structural Properties	Technical	The structural properties (lap-shear) pre- and post- exposure to corrosive environments is better or equal to requirements in industrial practices with conventional bonding techniques	SUCCESS!!
FY 15	Proven Efficiency	Technical	The NDE techniques used can prove the efficiency of the activation and re-assembly/bonding of the resulting joints	SUCCESS / In-Progress
FY 15	Characterization of Material Properties	Go / No-Go	The experimental characterization of material properties of the adhesive and adherend can be successfully performed to provide input to robust simulations (next phase)	SUCCESS / In-Progress GO ☑
FY16	Model Using Simulations	Technical	The simulations developed model the behavior and failure phenomena accurately without making crude assumptions and successfully agree with a wide range of experimental tests. <u>NOTE: Experimentally Validated Simulations!</u> An effort of 50% or more will be on experiments to validate and increase the robustness of the models, and to create reliable databases.	*In Progress* *Large-scale components & Environmental Testing*

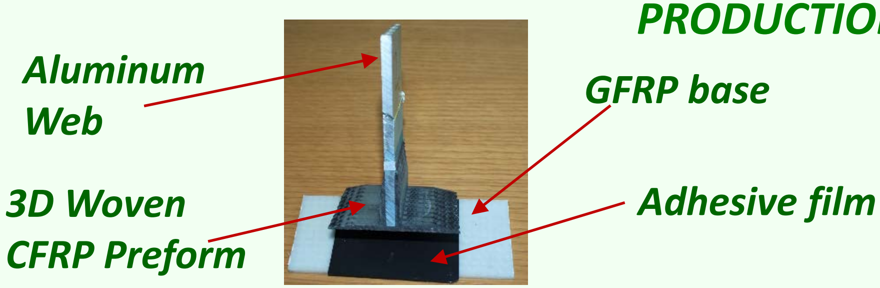
Progress: Active Adhesives – Film & Joint Production

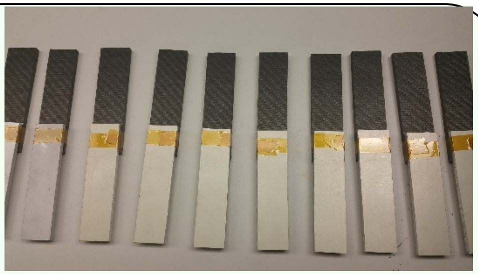
PRODUCTION OF ACTIVE ADHESIVES



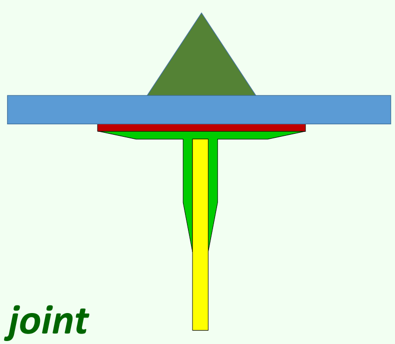
Thermoplastic *GnP* *Active Adhesive Pellets* *Injection molded discs* *Adhesive films + spacers*

PRODUCTION OF JOINTS

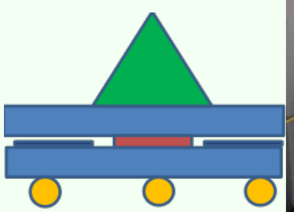


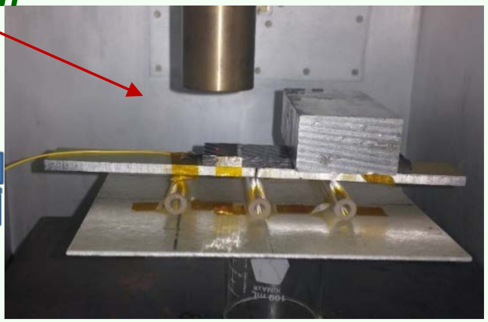


Microwave Horn



Microwave Horn





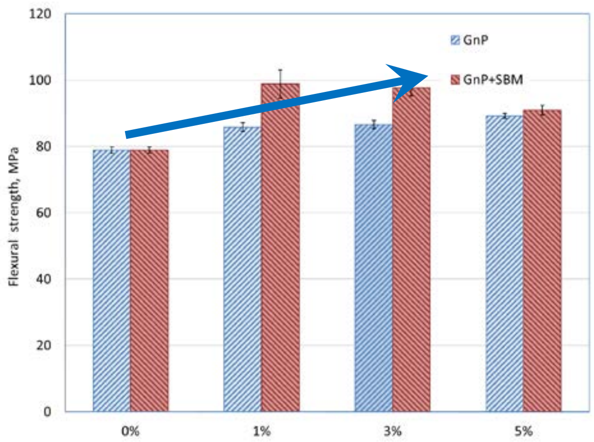
Single lap AL-CFRP joint

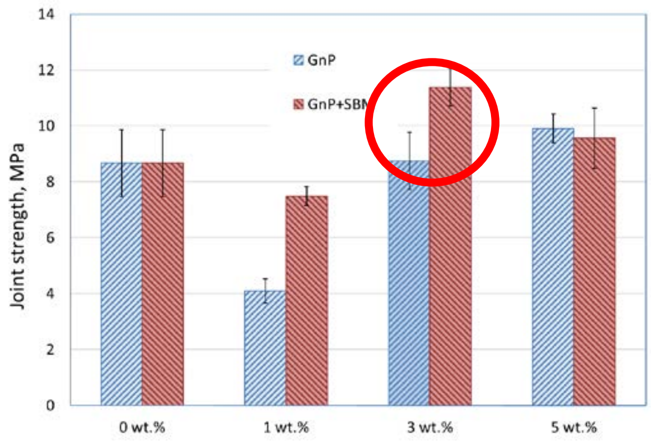
T- / Pi-joint AL-CFRP-GFRP joint

Progress: Technical Accomplishments/Results
Effect of GnP Functionalization

- **Functionalization of GnP:**
  - *Improve mechanical properties + Toughness + Multi-functionality*
  - *a) aliphatic epoxy (AE), b) phase separated elastomeric carboxy terminated butadiene nitrile rubber (CTBN), & c) styrene-butadiene-methyl-methacrylate (SBM) triblock polymer.*
  - *SBM functionalization has shown the greatest potential.*

Improvement in Adhesive Flexural Strength



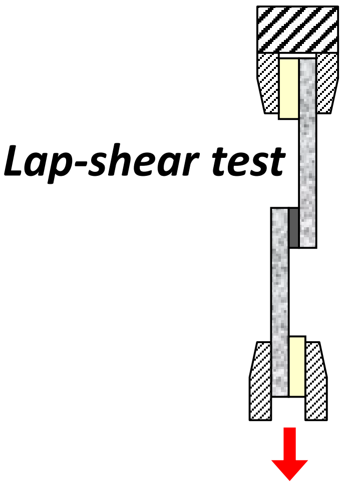


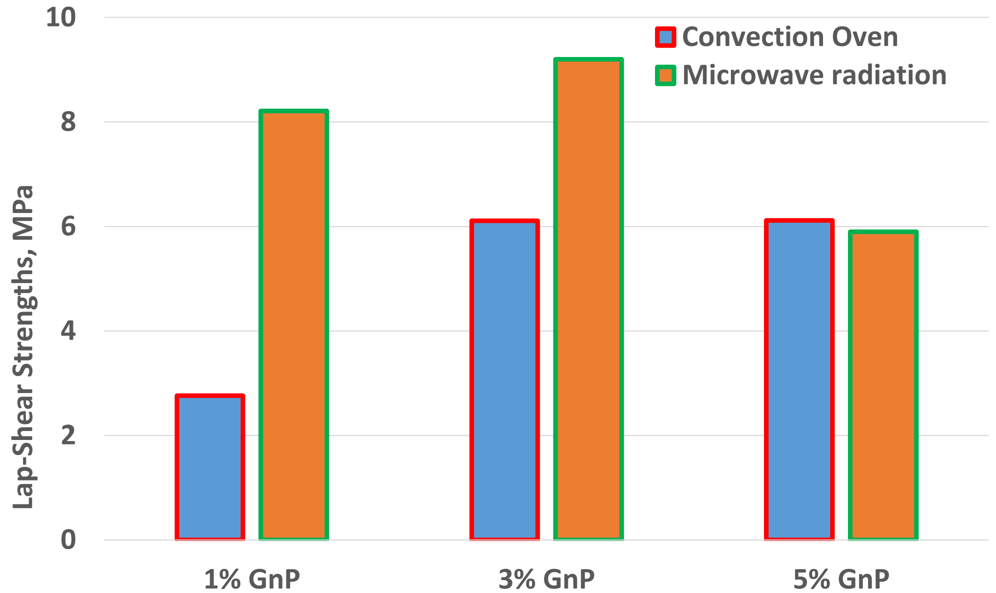
Improvement in Lap-joint Shear Strength

- *For 5 wt.% GnP , on average flexural strength of Nylon-6 was enhanced by > ~10%.*
- *For 1 wt.%, 3 wt.%, on average flexural strength of Nylon-6 enhanced by > ~20%.*
- *At 3 wt.%., the lap-shear strengths of GnP + SBM-functionalized adhesives were improved by **more than 30%** relative to pristine adhesives.*

Progress: Technical Accomplishments/Results Conventional Thermal Vs Microwave Bonding

➢ ***Lap-shear joints strengths:***



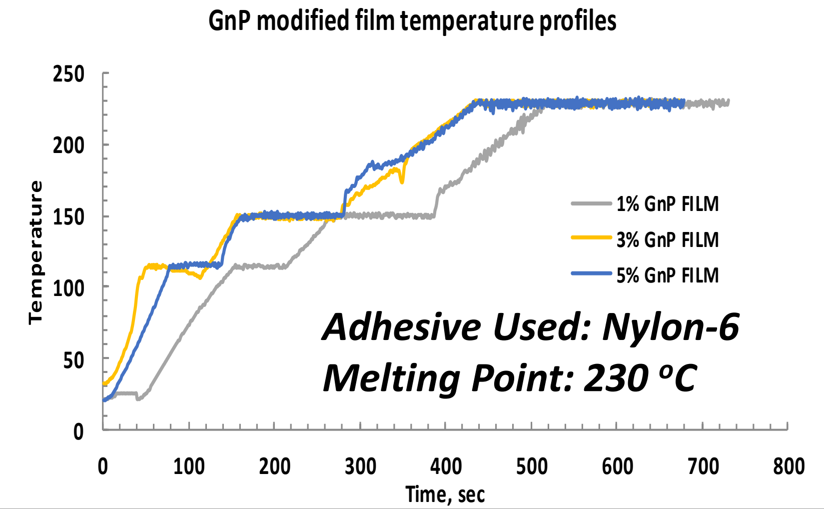


Microwave activated joints showed better performance. This could be due to several reasons.

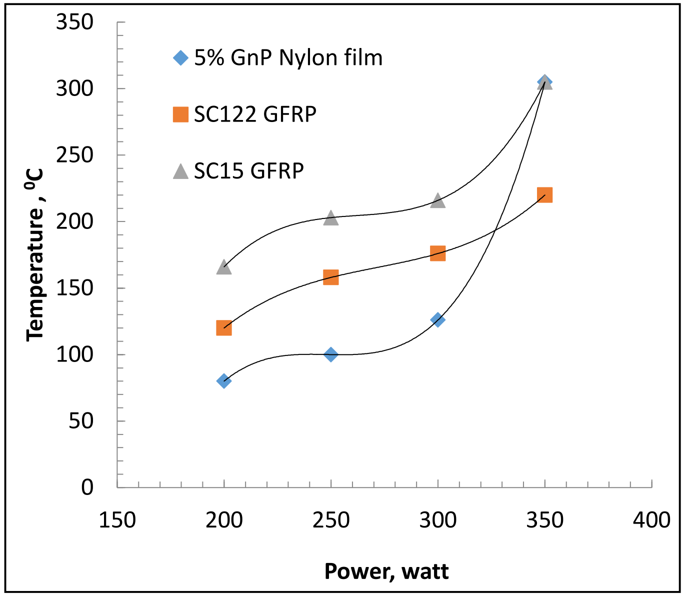
- ***Firstly, the adhesive heats up uniformly through out the bond-area. This may reduce the residual stresses developed in the adhesive and thereby increase the joint-strength.***
- ***Secondly, In thermally bonded systems, edges are heated first, as the heat is transferred via conduction from the substrates to the adhesives, and via convection through the edges of the adhesives, thereby degrading the adhesives at the edges and reducing strengths.***
- ***Lastly, in microwave assisted heating, the substrate does not degrade as the adhesive is heated rapidly and in most cases does not exceed the T_g of the substrate.***

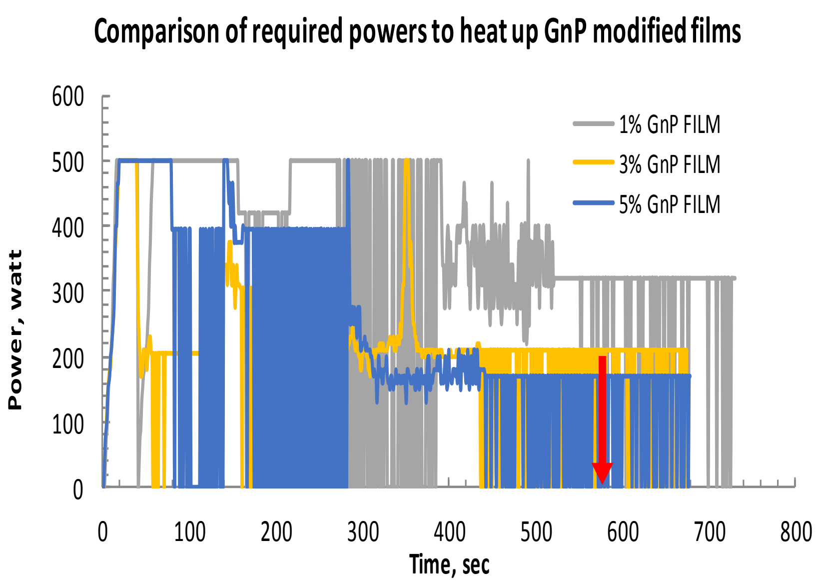
Progress: Technical Accomplishments/Results
Targeted Heating of Adhesives

- ***TOP RIGHT: 3 wt.% and 5wt.% GnP films heat faster***
- ***BOTTOM RIGHT: As the GnP content increases, the power required to reach target temperature reduces.***
- ***BOTTOM: Instead of constant temperature, constant power shows promise in rapid heating of adhesives relative to substrates. Experiments with embedded sensors in adhesive and composite substrates are in progress to corroborate these findings.***



Comparing temperature of adhesive and substrates at a constant power for a constant time



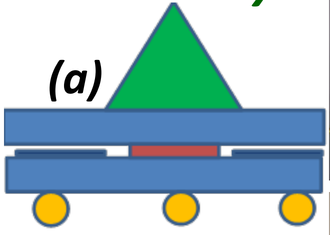


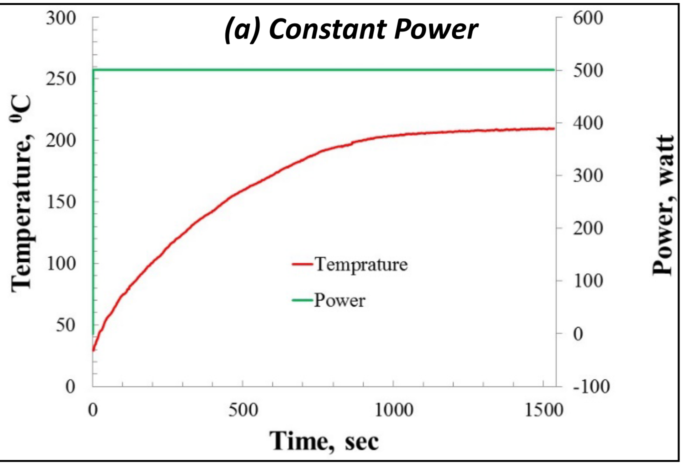
Progress: Technical Accomplishments/Results
Al - CFRP : Lap-Joint Assembly

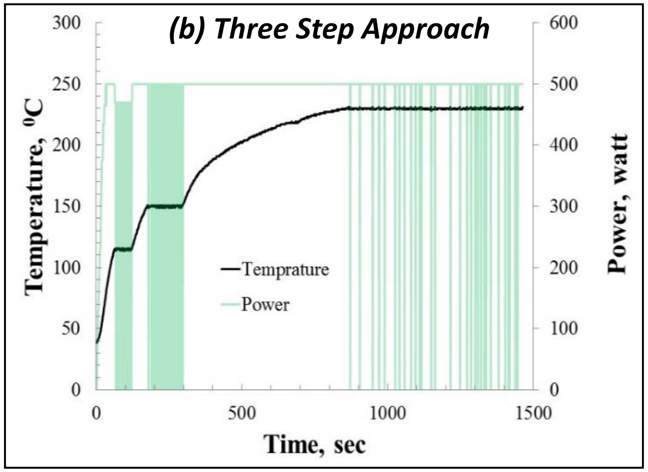
- ***Reversible Bonding of AL-CFRP Single Lap-Joint with Nylon6 + xGnP***

Approaches

- ***Two approaches: a) constant 500 watts and b) three step approach, temperature control, variable power.***







(a) Schematic of Bonding,
(b) real joints,
(c) Completed Joints.

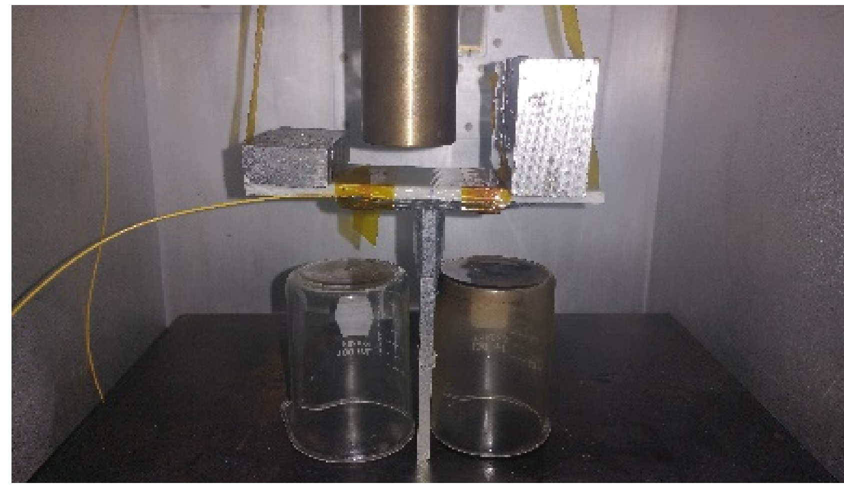
- ***Three steps VFM recipe exhibited rapid heating process relative to the constant power recipe, e.g., to reach at 150 °C,***
- ***The constant power recipe took about 445 secs, whereas the three steps VFM recipe took only about 180 secs. Plus we <u>have lower consumption of power</u>***
- ***In short, the process is <u>tailorable</u>! Depending on substrate, adhesive, GnP content and processing time required, it can be <u>designed</u> accordingly***

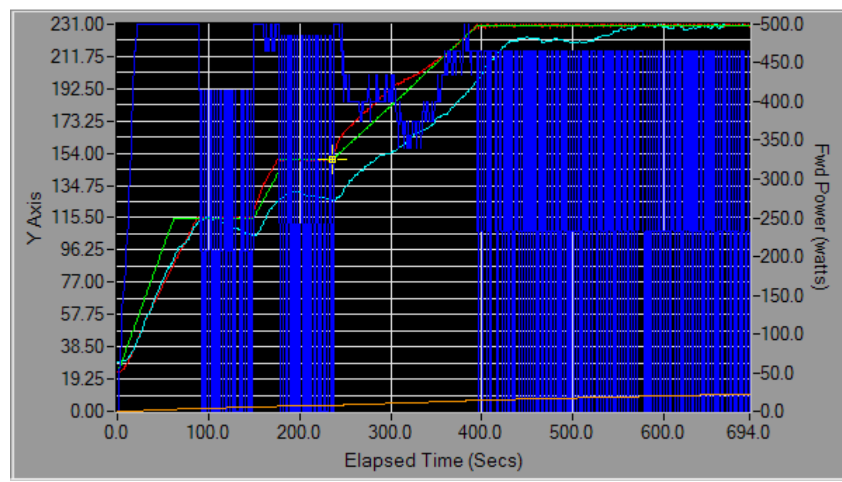
- ***Similar process carried out for disassembly***

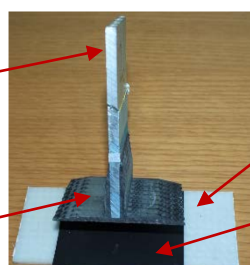
Progress: Technical Accomplishments/Results Dissimilar material Pi-/T- joints (out-of-plane)

➢ ***Reversible Bonding of multi-material T-/Pi-Joints with Nylon6 + xGnP***

- ***FOR BONDING**, each individual sample was placed inside the MC2100 VFM cavity.*
- ***Metallic block weighing ≈450 g, were used to provide process hold down weight.***
- ***Bottom right figure shows Temp. (red + green) vs. Power (blue) variations***
- ***GFRP substrates allowed rapid heating of adhesive ~ 230 °C within 400 s. This heating was slowed down with CFRP base. Further optimization is necessary.***
- ***Nevertheless, successful assembly, dis-assembly, and re-assembly carried out.***







Aluminum Web

GFRP base

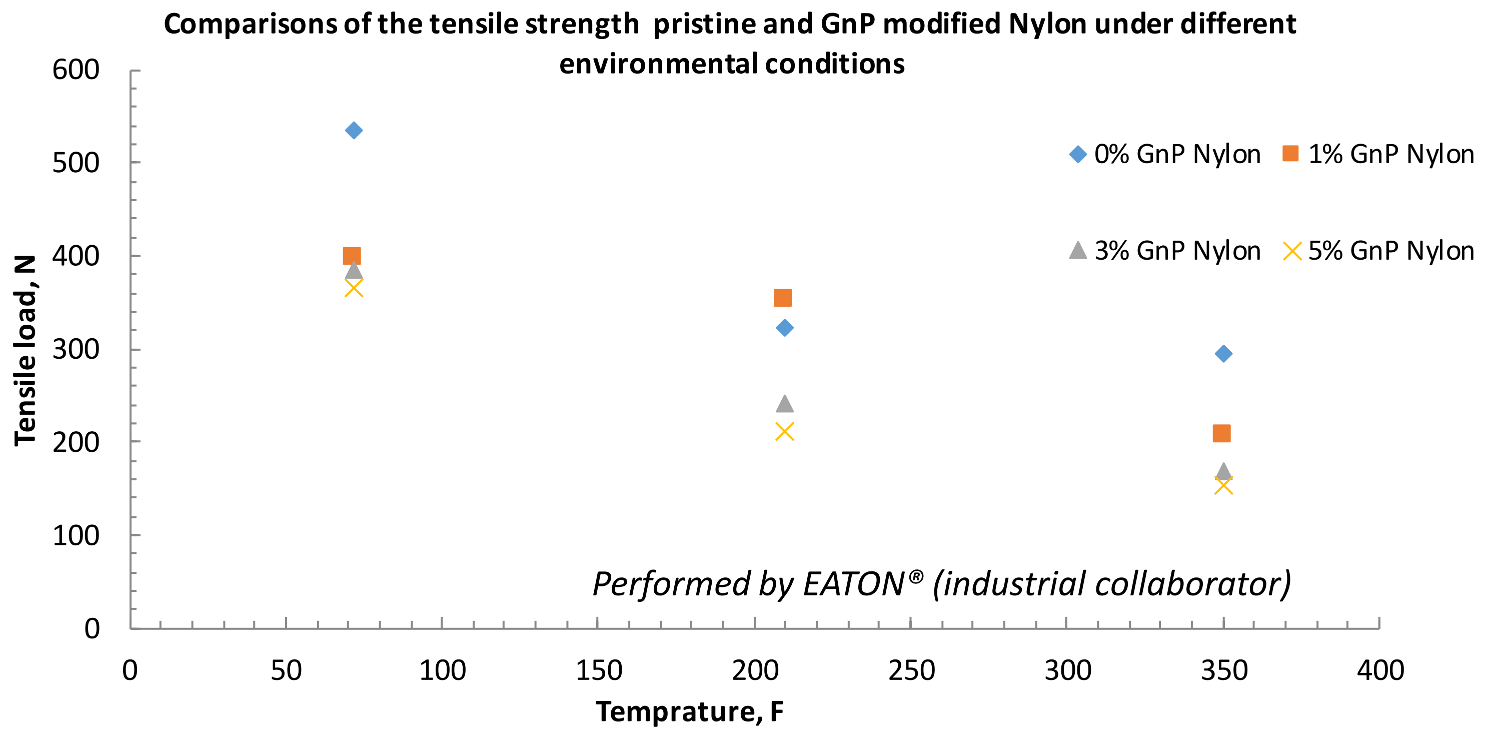
3D Woven CFRP Preform

Adhesive film

Progress: Technical Accomplishments/Results Testing at High Temperatures

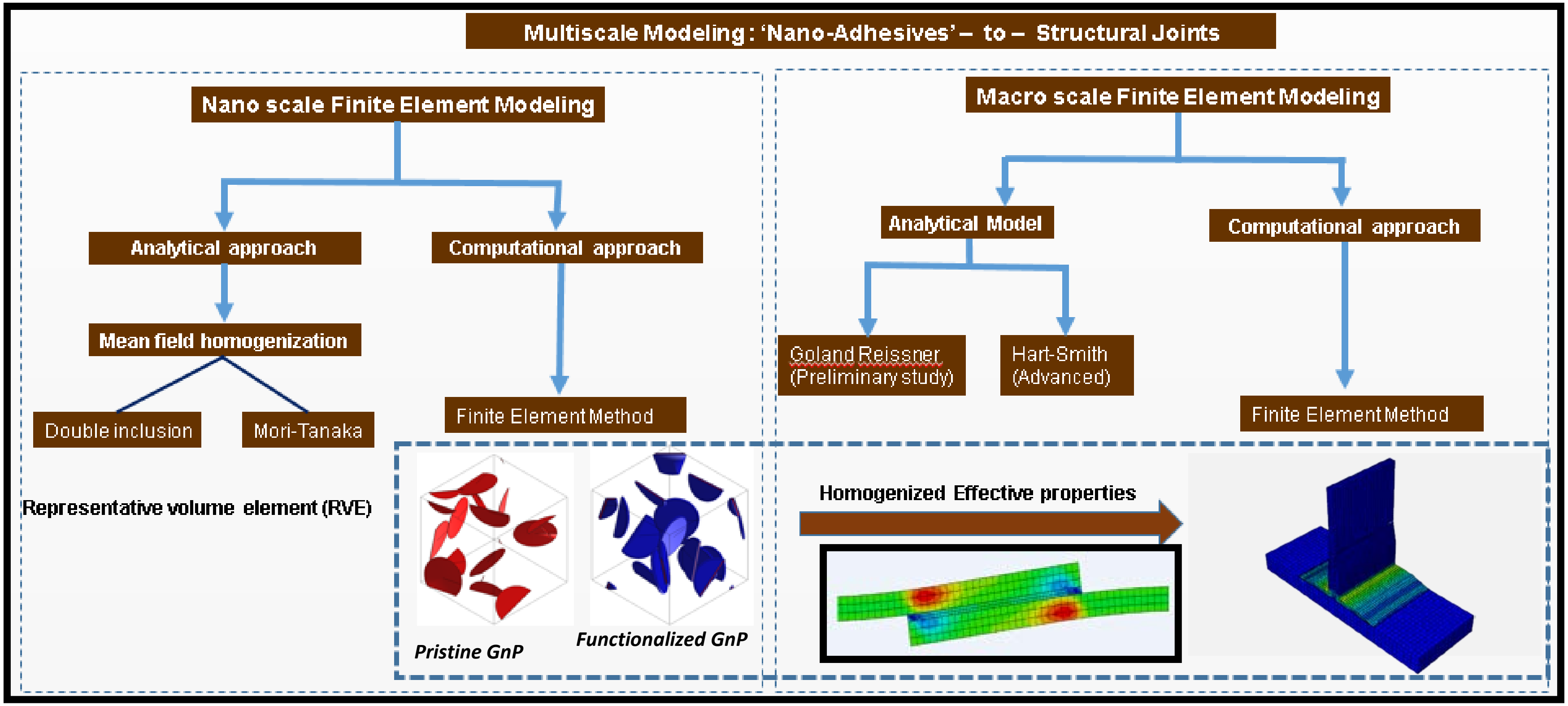
➢ ***Materials characterization under varying temperature***

Tensile test



- ✓ ***As expected, increase in temperature reduced the tensile strengths.***
- ✓ ***Increase in thermal conductivity of adhesive due to GnP may have contributed to further decline in tensile strengths at high temperatures. This was evident at 350 F wherein tensile strength dropped as GnP content increased.***

Progress: Technical Accomplishments/Results Overview & Approach in Modeling



Study of adhesive characterization

- Effective stiffness, toughness, thermal & electrical conductivity.

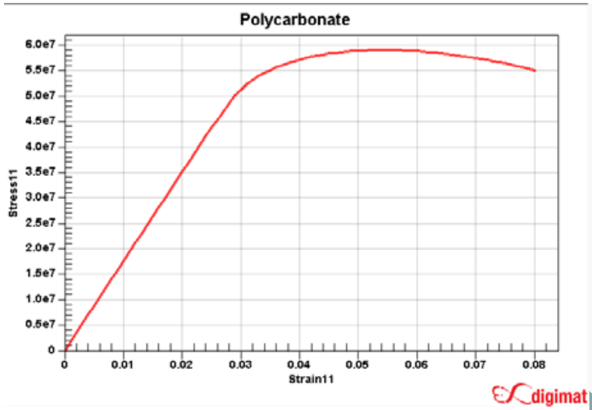
Study of Multi-material joining

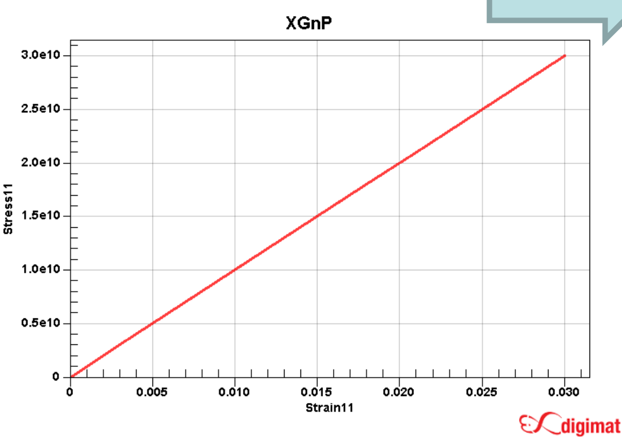
- Structural behavior, modeling damage/failure, progressive damage and development of experimentally validated simulations (EVS)

- ***Experimentally Validated Nano-scale models to help predict the structural behavior beyond the experimental matrix in this study***

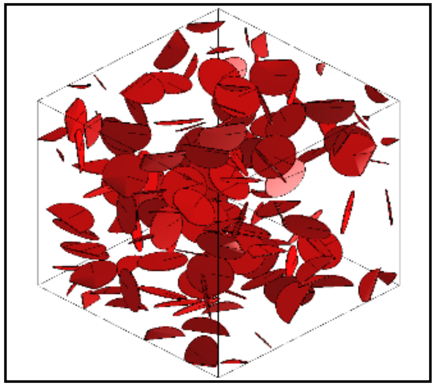
Progress: Technical Accomplishments/Results
Nano- / Micro- Scale Modeling

Material Models

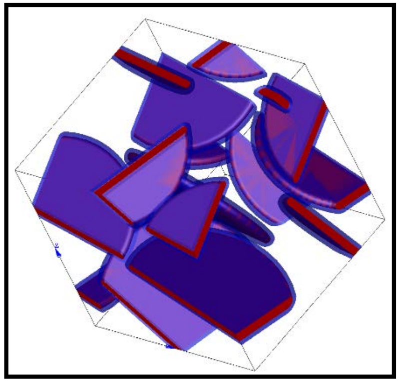




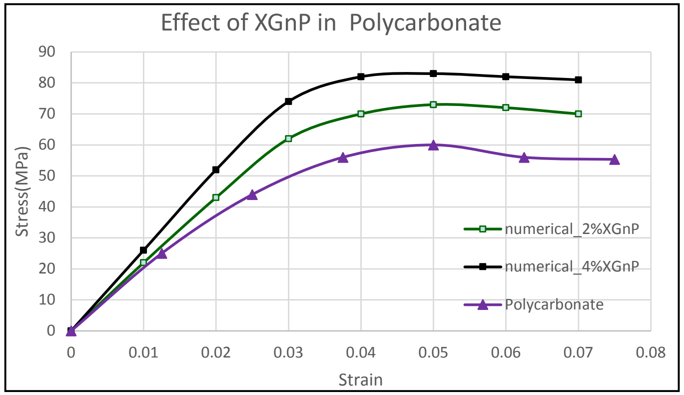
Unit Cell / Realistic MODELING



Microstructure of 2 wt.% Pristine GnP in Polycarbonate



Microstructure of 2 wt.% functionalized GnP in Polybonate

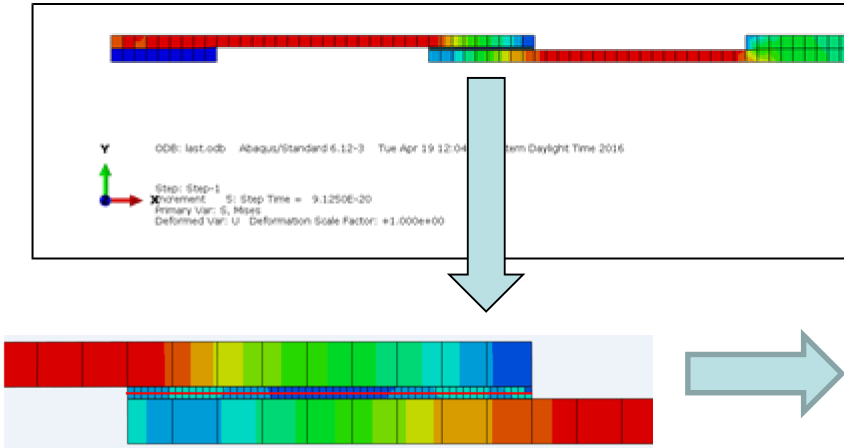


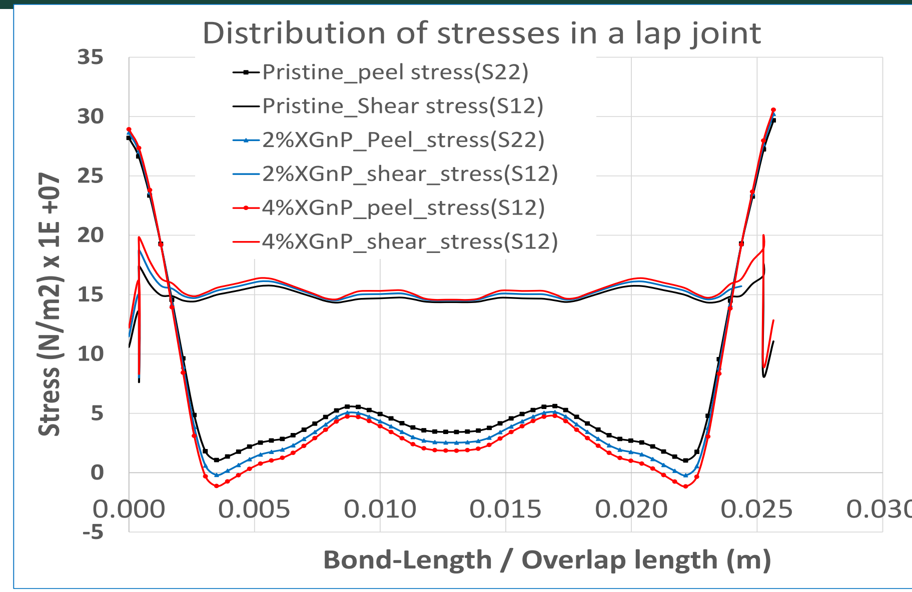
Prediction of Adhesive behavior with XGnP

- ***Realistic modeling and successful prediction of <u>nonlinear behavior</u>***
- ***Successful modeling of GnP/polymer interfaces to take functionalization into account.***
- ***Material model can be directly <u>input to structural models</u> or can <u>linked as multi-level models</u>***

Progress: Technical Accomplishments/Results
Numerical Modeling – Lap Joint

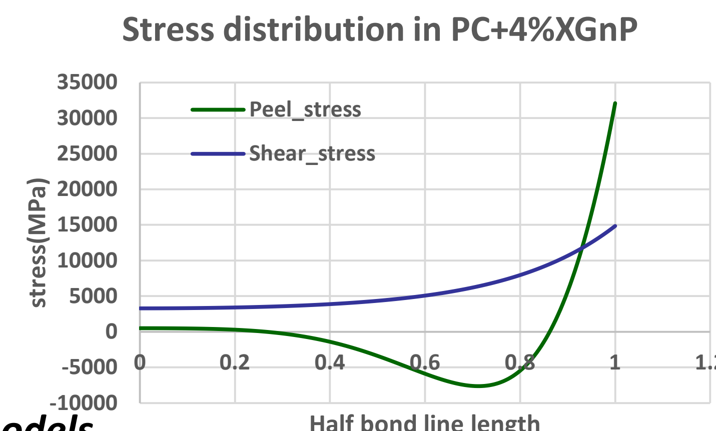
- ***Adhesive Material Model from Nano-scale input into structural models***





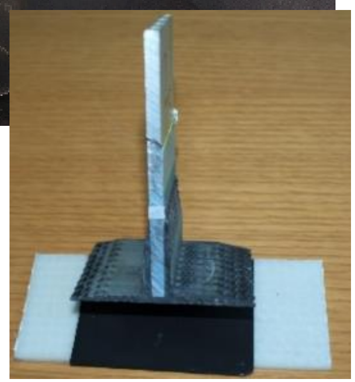
- ***GnP increases stiffness of adhesives***
- ***Clear effect on reduction of peel stresses.***

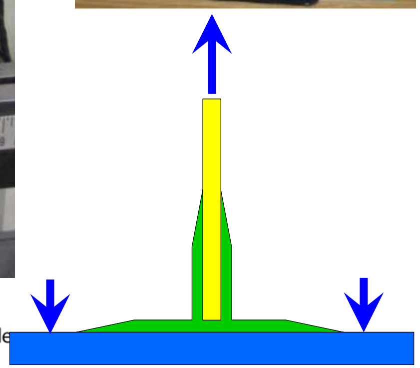
- ***Analytical Models:***
  - ***To be used as thumb-rule / conservative cut.***
  - ***Plot on right: Goland-Reissner model***
    Key <u>Assumptions:</u> substrates are thin-beams
  - ***Future work will include Hart-Smith and advanced models that include tip-plasticity.***

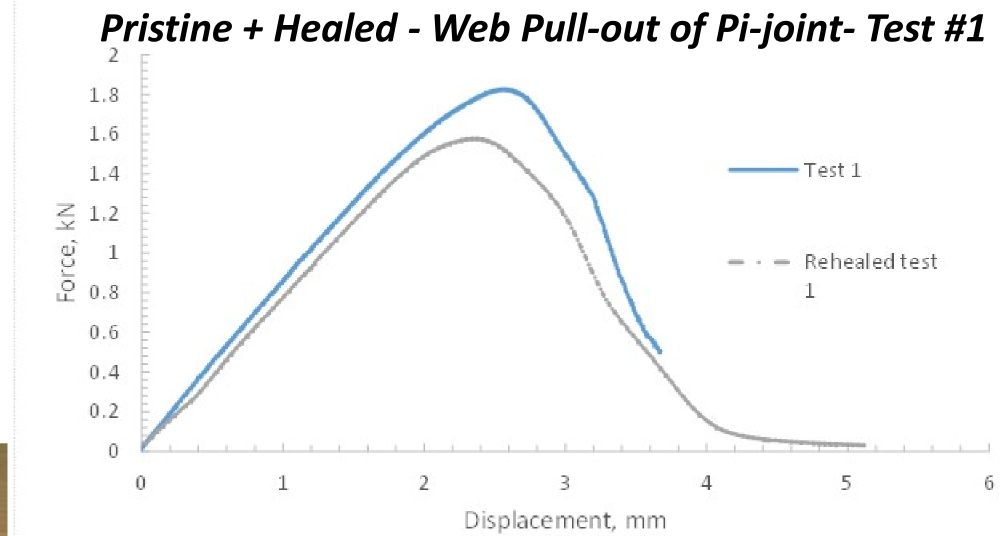


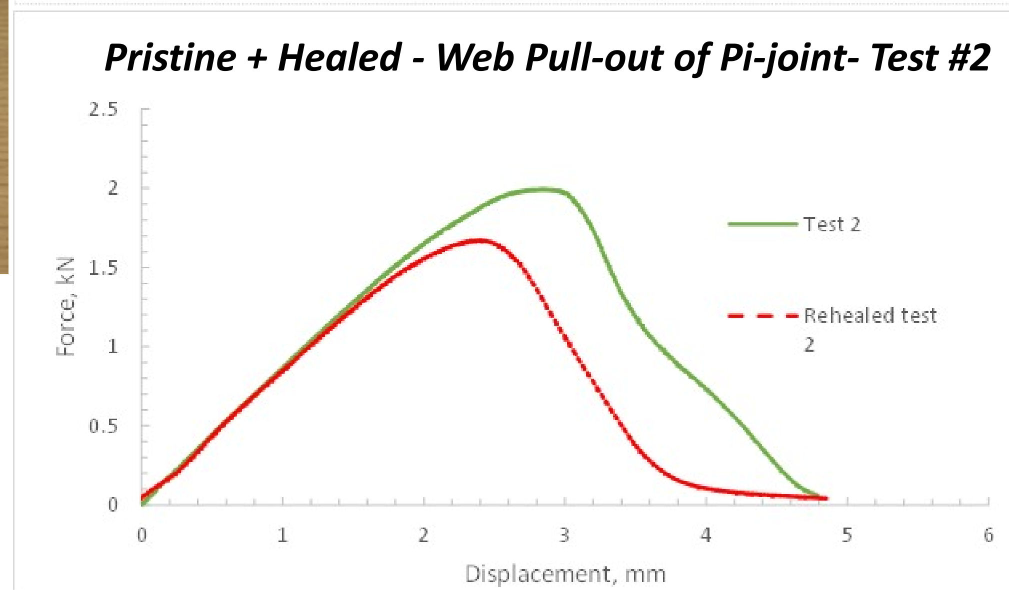
Progress: Technical Accomplishments/Results Prototype –Assembly/Dis-assembly/Healing

ASSEMBLY + TESTING + HEALING



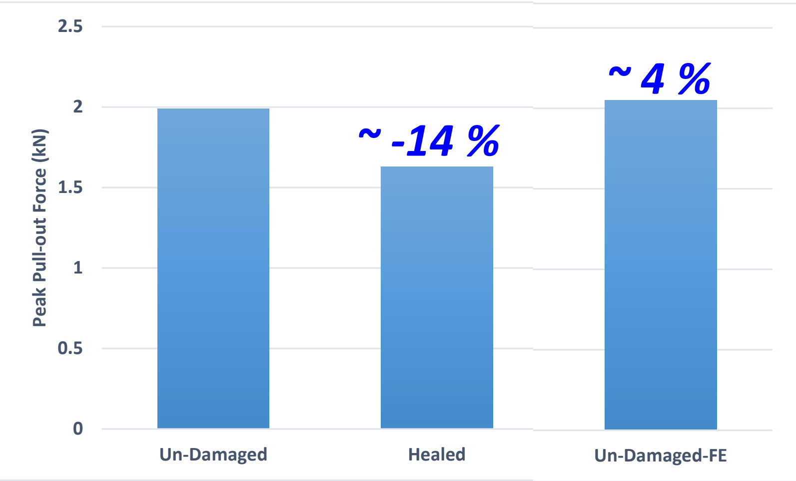






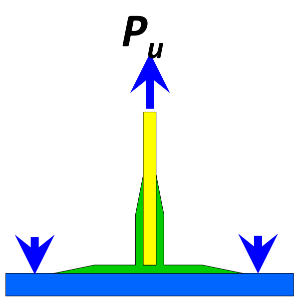
MICHIGAN STATE UNIVERSITY | Composite Vehicle Research Center

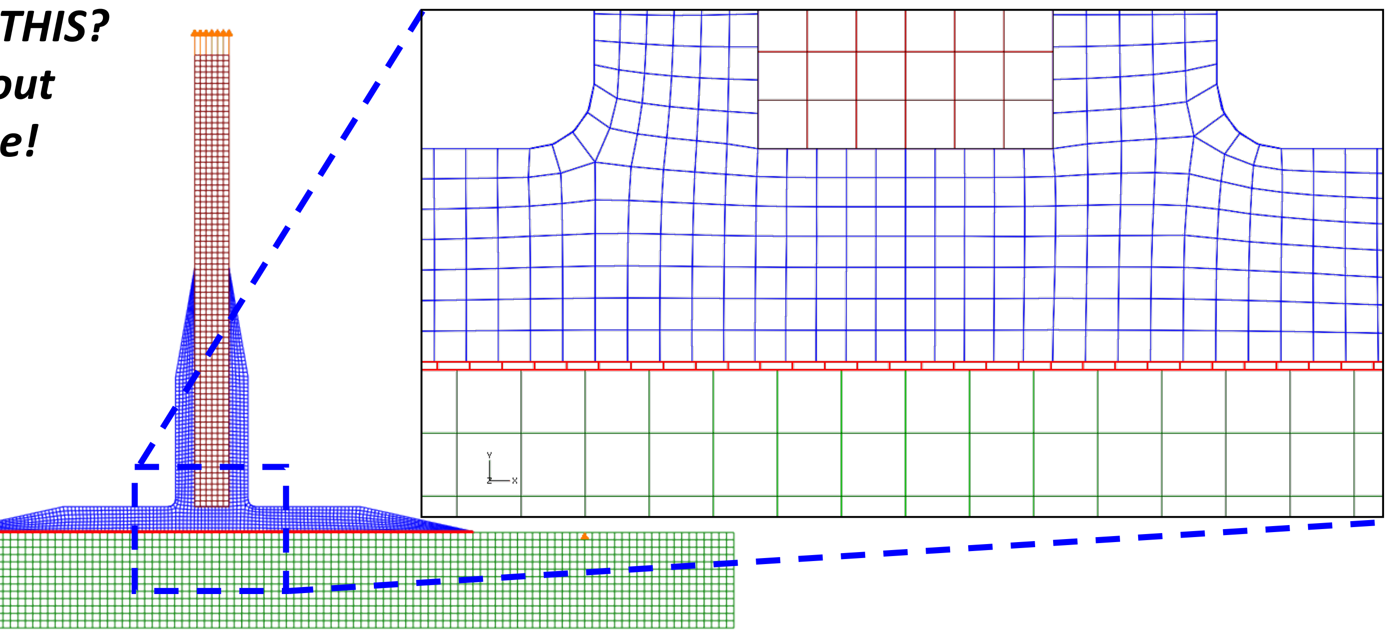
Progress: Technical Accomplishments/Results
Simulations Vs Experiments



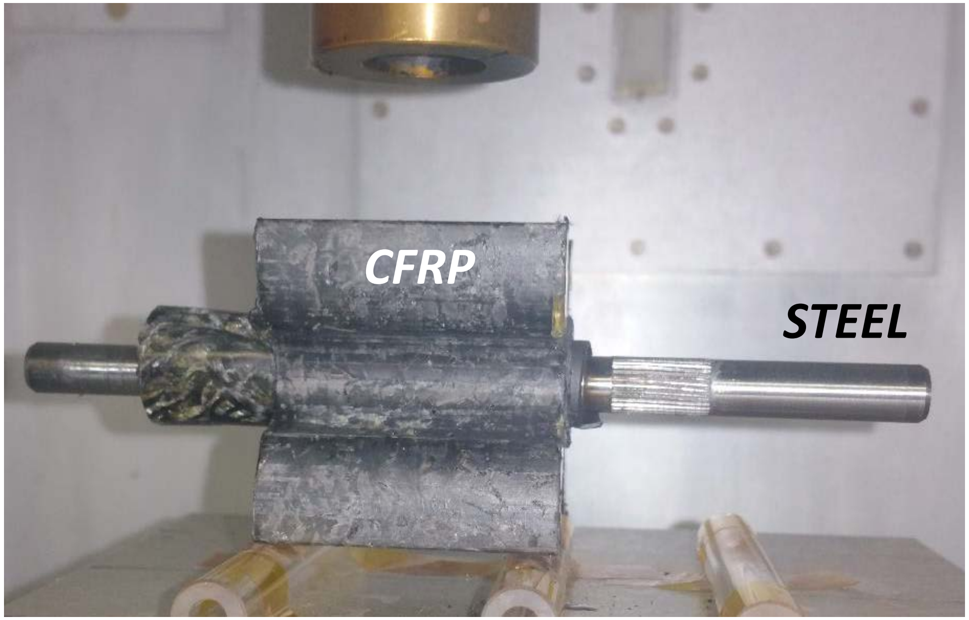
- **2D , plane stress models were simulated in ABAQUS®**
  - The adhesive was modeled with a finer mesh
  - Allows for detailed modeling of flaws
- ***Use Experimentally Validated Simulations***
- ***Predict Behavior of All possible damage locations***
- ***Obtain a Design Space, 3D - Performance Surfaces!***
- ***Develop Design Charts for easy use (in the field !)***

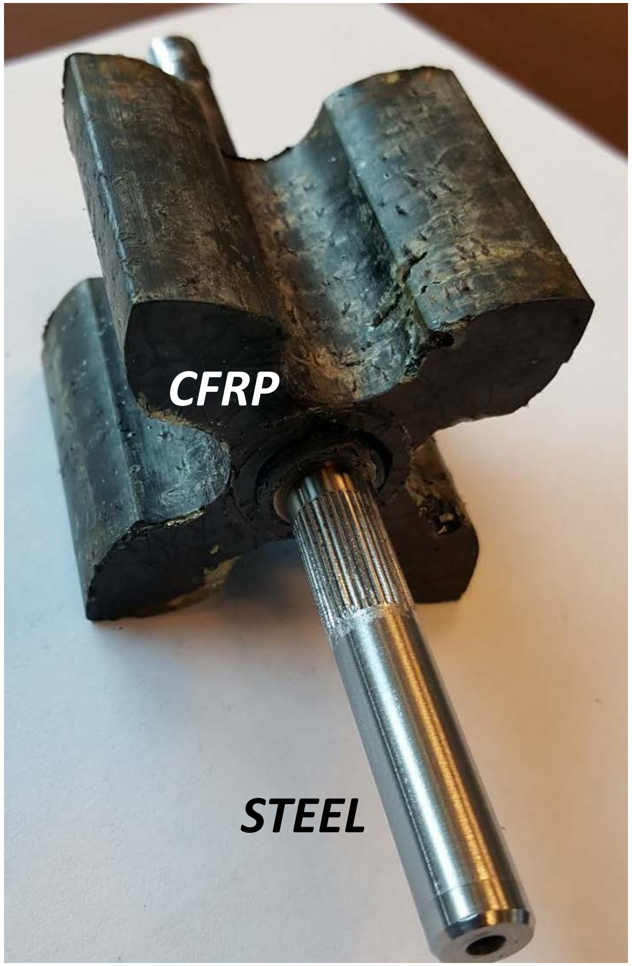
Can We Simulate THIS? With and without Flaws/Damage!





Progress: Technical Accomplishments/Results Prototype – Activation –Assembly + Healing





TOP: Microwave activated joining
RIGHT: Assembled /Bonded rotor

- ***Commercially available polyolefin based Thermoplastic (PRODAS 1400 hot-melt) used with 3 wt.% GnP***
- ***Successful Activation and Bonding of Steel shaft to CFRP rotor***

Response to Previous Year Reviewers' Comments

Reviewers' Comment	Action Taken and Results
#1. The reviewer liked the way the project is advancing and hopes the present momentum can be maintained to the end of the project.	The investigators have used the lessons learned from the first and second year as a launch-pad and further carried the momentum to include prototypes of dissimilar materials including CFRP, Al, Steel. Further healing/recovering in-service degradation has been proved.
#2. The reviewer judged that the approach as not clearly appropriate for this study and questioned exactly how the investigators will use a rational computational materials approach to advance this study. The reviewer observed that no evidence is given in this presentation, and said that there is an apparent random walk rather than a directed approach.	The reviewers' observation and comments are accurate and appreciated. At the end of the last review, experimental validation and development of multi-level numerical models were in progress. In the current performance period considerable progress in development of nano-, meso- and macro-scale models has been performed, and a modeling scheme that predicts structural joint behavior from nano-scale to structural level has been developed. Plus, novel NDE tools have been used to further increase the robustness of the developed models.
#3. The reviewer cited good results but offered it would be better to use an adhesive other than nylon, because the auto industry makes only limited use of nylon due to its affinity for moisture.	Agree. The investigators have used nylon-6, polycarbonate and a commercially used automotive adhesive (Prodas 1400). Additionally, with recommendations from program managers and industry input, ABS will also be evaluated in the rest of the performance period.
#4. The reviewer stated resources were insufficient, recommending the team should include the current car industry participation and also add other industries where bonding is a significant part of their businesses.	Agree. This project has gained considerable attention from industry. Invited talks and conference presentations have paved the way for exchange of ideas and communications. While the car industry is directly not involved in this work at the moment, their input is being incorporated in this project and future work will directly involve them.

Collaborations & Coordination

Collaborators / Partners	Details
Eaton Corporate Research and Technology **(PARTNER)**	✓ Low-inertia, light-weight, supercharger applications ✓ High-speed rotational/torsional testing ✓ Non-destructive Evaluation at high speeds ✓ Metal – to- metal and Metal to composite Bonding ✓ In-situ repair, assembly and disassembly
U.S. Army TARDEC **(In-kind Collaborator)**	✓ Periodic review of progress and guidance on relevant materials for automotive applications and path forward.
OakRidge National Laboratory **(ORNL)**, Carbon Fiber Technology Facility **(CFTF):** **(In-Kind Collaborator)**	✓ Low-cost, Large-Tow Carbon Fiber. ✓ Guidance of possible automotive applications

Barriers and Solutions + Future Work

- **CHALLENGES / BARRIERS:**
- **Semi-crystalline thermoplastics are very susceptible to processing parameters, specifically mold temperatures and can lead to high scatter in resulting structural properties. ADDRESSED: Amorphous thermoplastics and consistent processing methods have shown promise. Also, multiple thermoplastics are being explored.**
- **Microwave Equipment: The sample size is still limited by the size of the VFMW oven. POSSIBLE SOLUTION*: Collaboration with Lambda Technologies has revealed the possibility of a field applicator that can be placed on a robot arm for field applications.***

- ***FUTURE WORK (Current Budget-Period):***
- **Corrosion Analysis followed by structural testing.**
- **Continuous optimization and narrowing down of processing parameters**
- **Statistically significant testing at both room and elevated temperature of all multi-material joints: a) In-plane, b) Out-of-plane, & c) Torsion**
- **Re-assembly and In-situ Repair (post-fatigue and post-impact).**
- **Non-Destructive Evaluation: a) Guided Waves, b) IR Thermography, & c) Fiber-optic sensors.**
- **Dissemination of Results**

Summary

- **RELEVANCE:**
  - Joining & Assembly: Multi-material Joints that inherit the benefit of both bonded (lightweight) & bolted (re-assembly+repair) joints through 'active,' 'reversible,' adhesives.
- **APPROACH:**
  - Reinforcement of thermoplastic adhesive with novel graphene nano-platelets (GnP) and to use GnP/microwave-interaction for 'targeted heating of adhesive' thereby allowing ease of repair and re-assembly
  - An Integrated Experimental & Simulations based approach that eliminates the trial-and-error approach is adopted. Robust design tools are also developed.
- **KEY TECHNICAL ACCOMPLISHMENTS**
  - Targeted heating of adhesives, dis-bonding and re-assembly and "Healing" was proved Multi-materials, various adhesive and three types of joints successfully developed.
  - Numerical simulations at nano-, meso- and macro-scale developed and experimentally validated. A multi-level scheme that can predict the structural behavior by taking into affect the nano-particle distribution developed.
- **Partners / Collaborations:** Eaton Innovation Center, MI.
- **FUTURE WORK:**
  - Corrosion and Elevated Temperature testing
  - Further optimization of processing parameters
  - NDE + Modeling +Development of Design Tools
  - Dissemination of Results and Findings

Active, Tailorable Adhesives for Dissimilar Material Bonding, Repair and Assembly

TECHNICAL BACKUP SLIDES

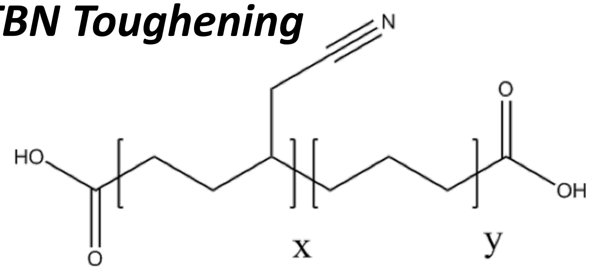
Progress: Functionalization of GnP

- **Functionalization of GnP:**

➢ Improve mechanical properties + Toughness + Multi-functionality

➢ a) aliphatic epoxy (AE), b) phase separated elastomeric carboxy terminated butadiene nitrile rubber (CTBN) , and c) styrene-butadiene-methyl-methacrylate (SBM) triblock polymer have shown the greatest potential.

CTBN Toughening



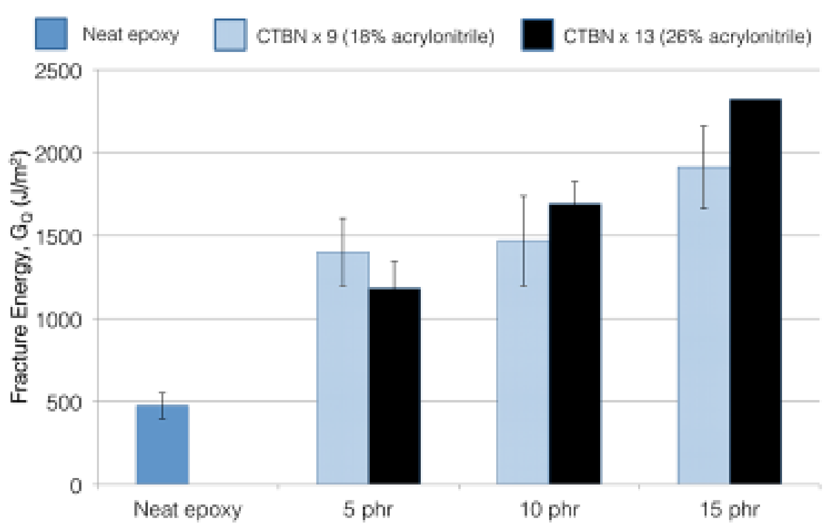
CTBN-carboxyl terminated butadiene acrylonitrile

➢ For brevity, only CTBN shown here.

➢ Functionalization of GnP with all three types of grafting (AE, CTBN, SBM) has been completed.

➢ Experimental characterization of multiple properties in progress.

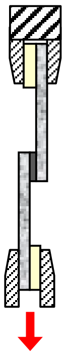
CTBN (18 and 26% acrylonitrile) in Epoxy: Fracture Energy (J/m^2)

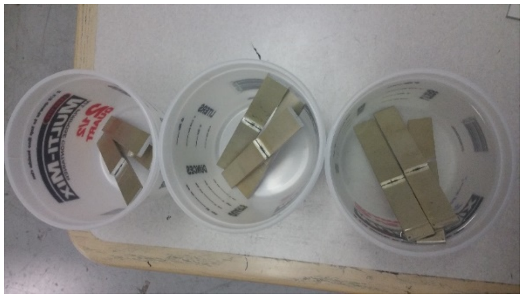


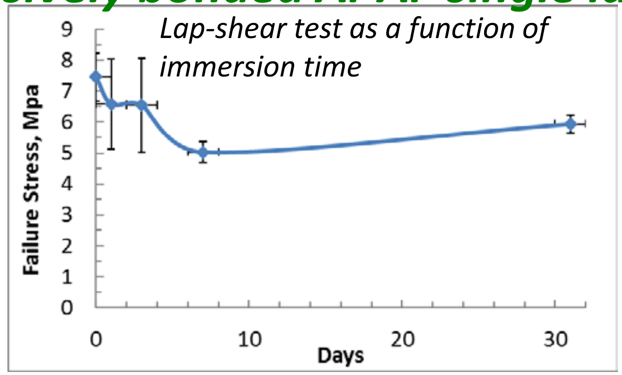
- G_{Ic} increases with increasing concentration of CTBN
- Maximum increase (400%) in G_Q was observed at 15 phr CTBNx13
- Increased acrylonitrile in the rubber additive leads to smaller particles and larger increases in fracture energy

Progress: Technical Accomplishments/Results
Environmental / Corrosion Testing

➢ *Water immersion tests for adhesively bonded Al-Al single lap joint*





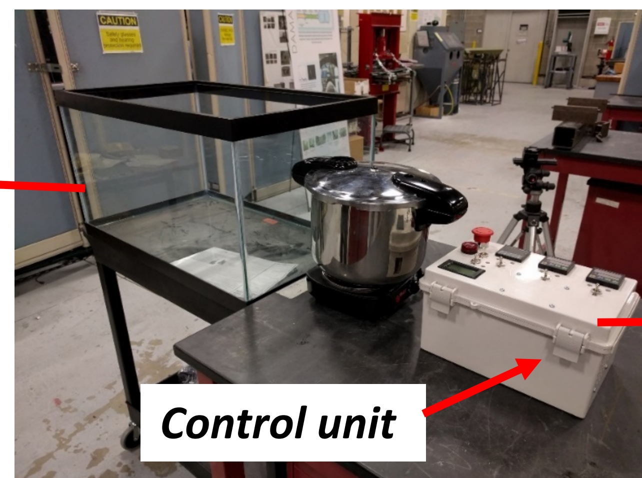


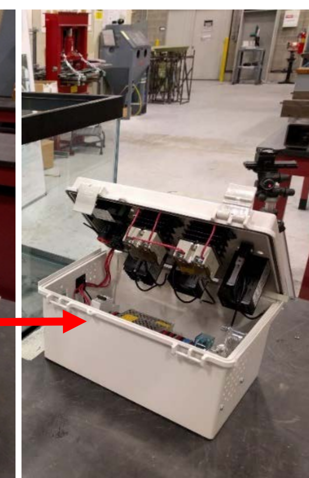
- *The strength of the single lap joints with Nylon-6 reduced by ~ 34% at steady state*

➢ *Development of environmental chamber for corrosion test of the Joints*

- ***The environmental chamber, which consists of Salt Spray (Fog) apparatus, is under development to perform the corrosion tests according to ASTM B117-11.***

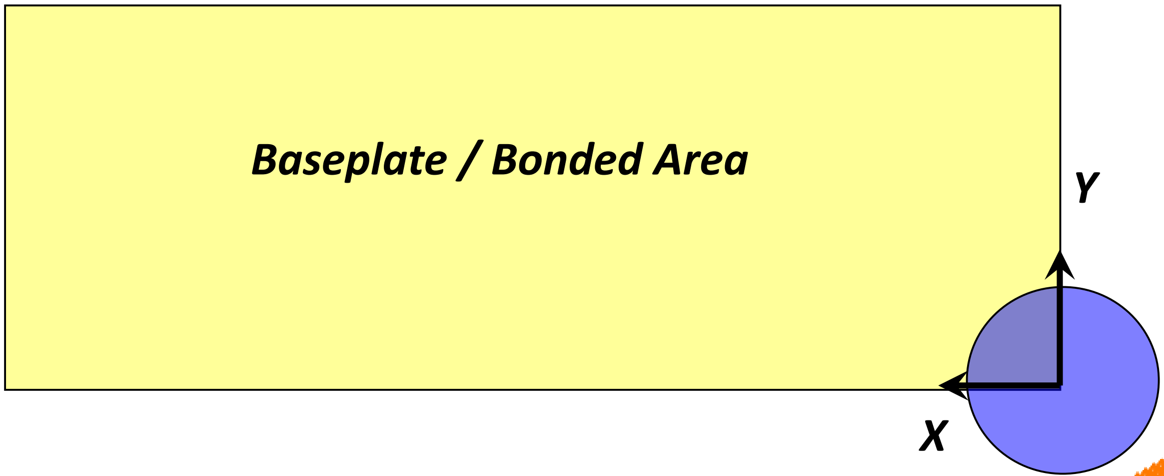
CAD model

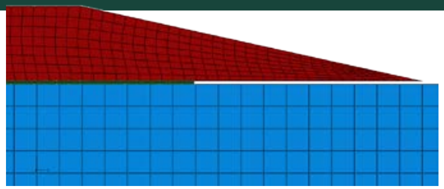




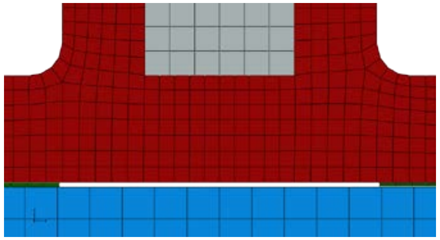
- ***Further environmental tests will be performed in this chamber.***

DESIGN TOOL & 3D Simulations



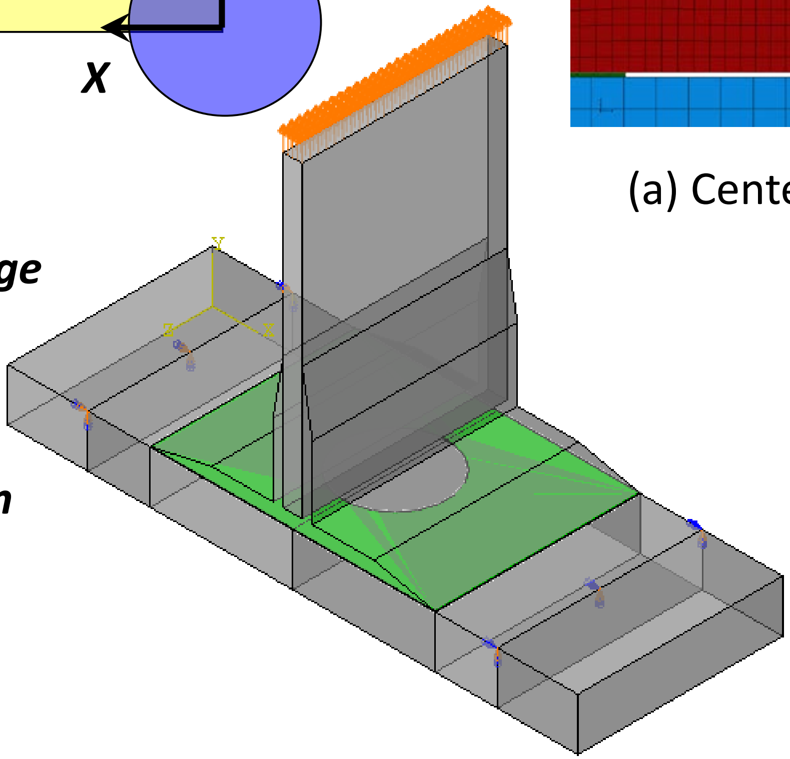


(a) Edge/Right Disbond



(a) Center Disbond

- ***Use Experimentally Validated Simulations (including flaws)***
- ***Predict Behavior of All possible damage locations***
- ***Obtain a Design Space , 3D - Performance Surfaces!***
- ***Develop Design Charts for easy use (in the field !)***



Progress: Technical Accomplishments/Results
DESIGN TOOL & 3D Simulations