



Enhancing Corrosion Resistance and Prediction of Corrosion Performance in Multimaterial Joints for Automotive Lightweighting Applications

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Richland, WA, USA**



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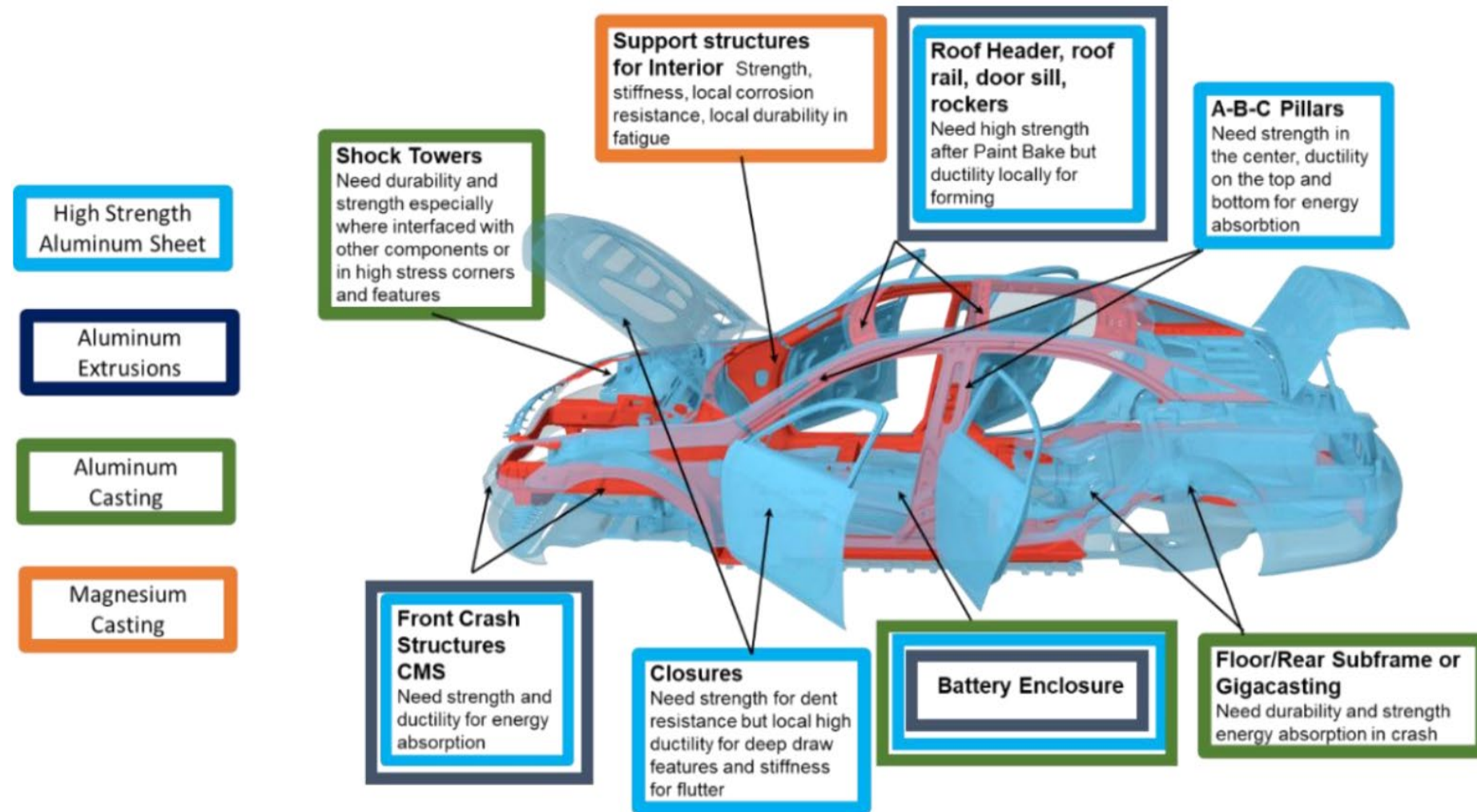


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We would like to thank the U.S. Department of Energy, Transportations Technologies Office (TTO), specifically Chris Schooler and Felix Wu for sponsoring this research as a part of the Lightweight Metals Core Program and the Joining Core Program.

The Pacific Northwest National Laboratory is operated by the Battelle Memorial Institute for the United States Department of Energy under contract DE-AC06-76LO1830.

Motivation



- Vehicle lightweighting, reduced part counts and lowered assembly costs.
- Corrosion in dissimilar materials joints is a key technical barrier to the successful integration of lightweight materials.

APPProach to Tackling Corrosion Challenges

Predict

Measure locally → Model physics → Predict risk

Drivers: Electrochemistry and microstructure

Method: SECCM / syringe cell → COMSOL

Target: Corrosion hot-spots & corrosion current map (where attack starts, how it evolves)

Prevent

(At the Joint)

Reduce driving force → Improve longevity

Drivers: Joint architecture & dissimilar metal contacts & crevice paths

Method: High-velocity coating to mitigate galvanic coupling / electrolyte access

Target: Reduced galvanic current & delayed initiation at joints

Protect

(At the Substrate)

Simulate & Optimize coating → Extend lifetime

Drivers: Reactive ligand substrate & defect sensitive coating stacks

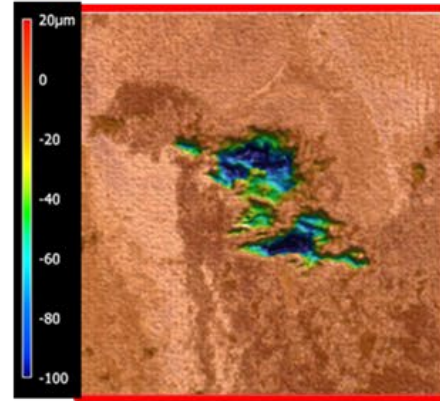
Method: Zn-rich spray coating to enable barrier/sacrificial behavior

Target: Improved coating durability & longer time to corrosion

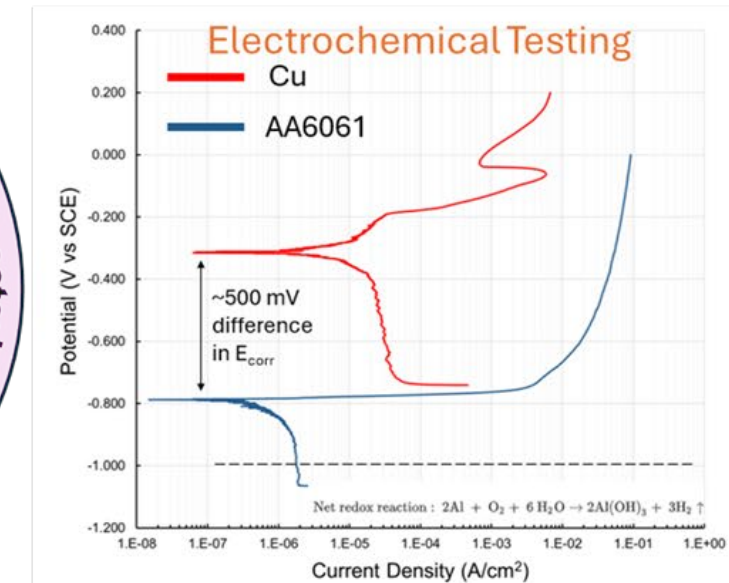
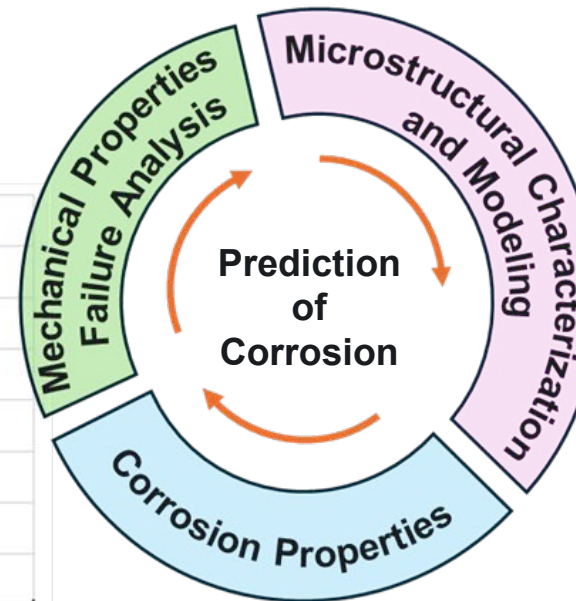
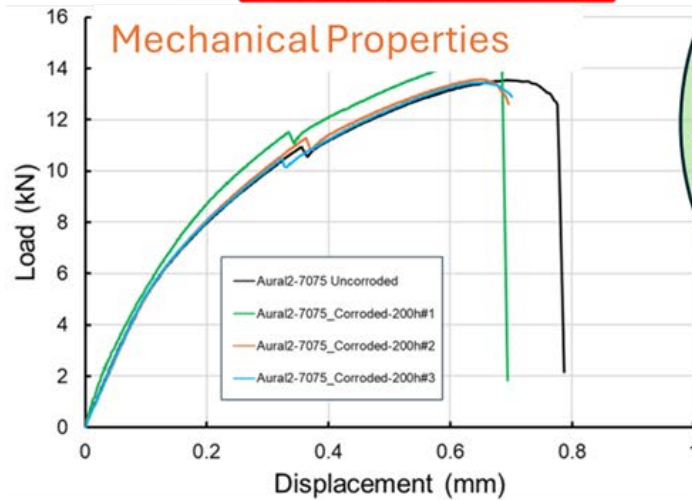
Prediction of Corrosion: Multimodal and Multiscale

Modeling and Experiments

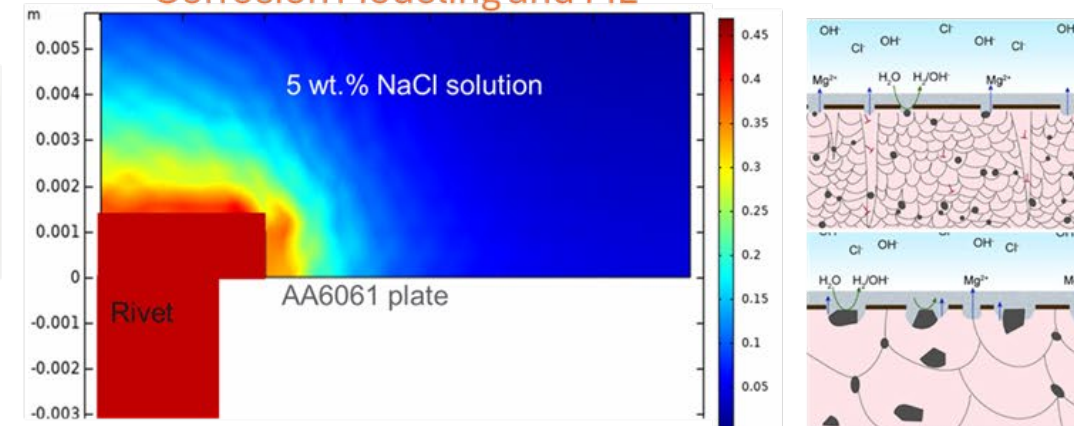
Failure Analysis Corrosion Pitting



As-fabricated joint samples



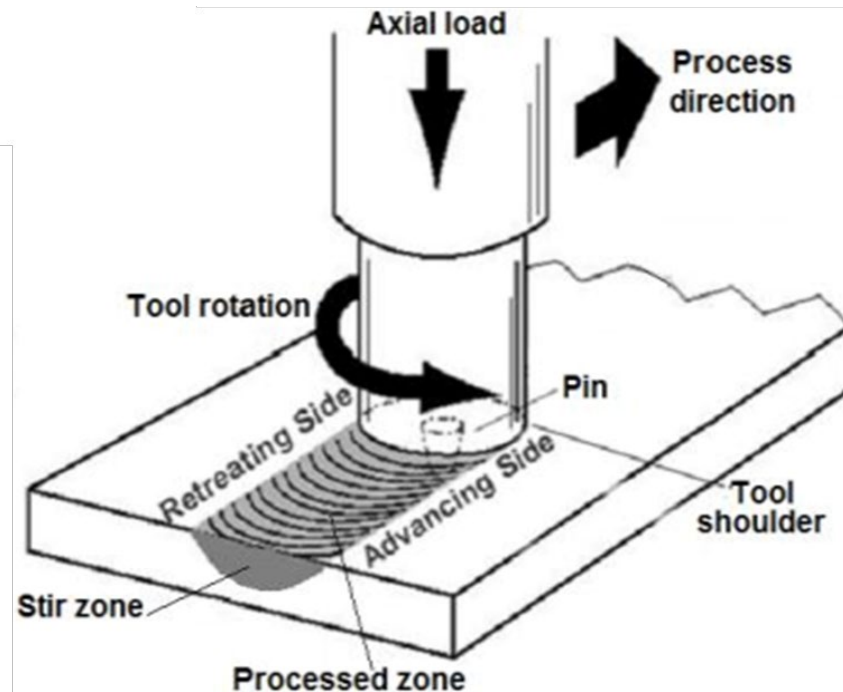
Corrosion Modeling and ML



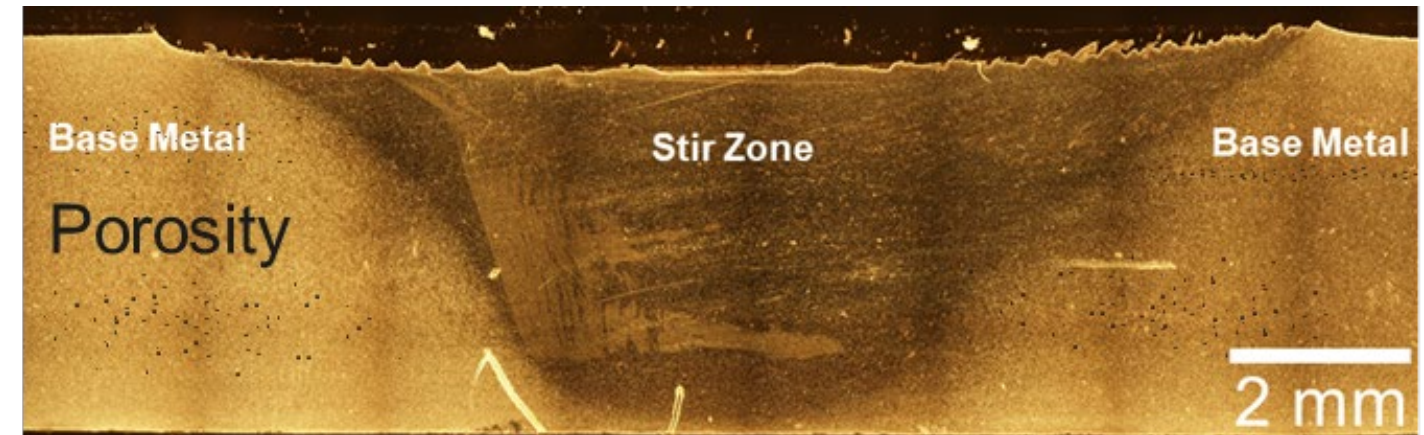
After Corrosion (ASTM B117- 200h)
Long term (days-weeks-months) Testing

Friction Stir Processing: Benefits and Challenges

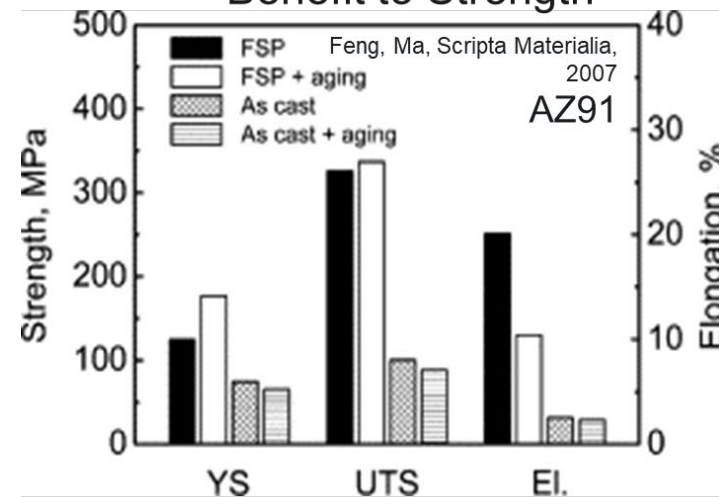
FSP improves localized material properties but introduces new corrosion challenges



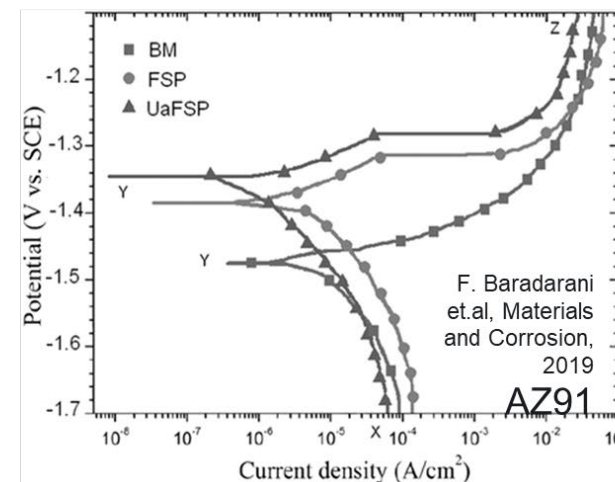
Etched Microstructure of FSP AZ91



Benefit to Strength



Benefit to corrosion



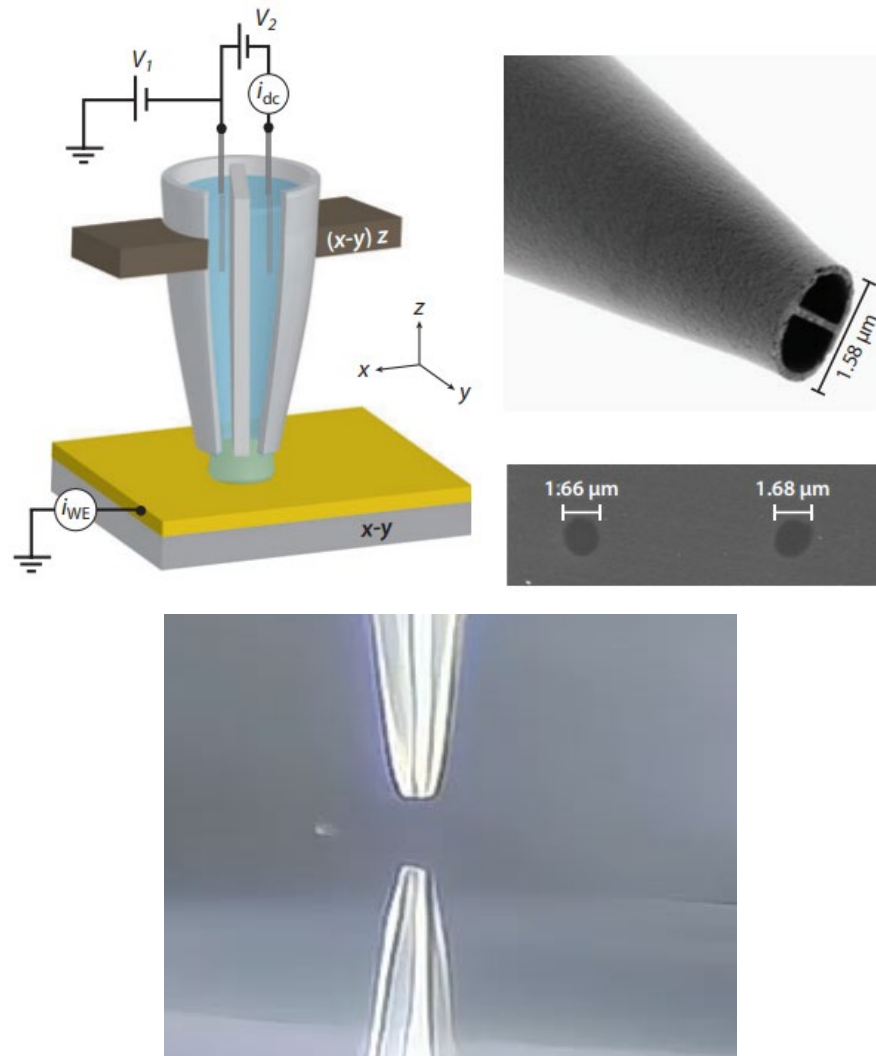
- Solid phase processing- No deleterious IMC formation.
- Targeted and localized microstructural enhancement- DRX and homogenization.
- FSP enhances both mechanical and corrosion properties within the stir zone.
- Corrosion properties of interface between stir zone and base metal = knowledge gap.

Investigating the interface between stir zone and base metal is critical to corrosion performance

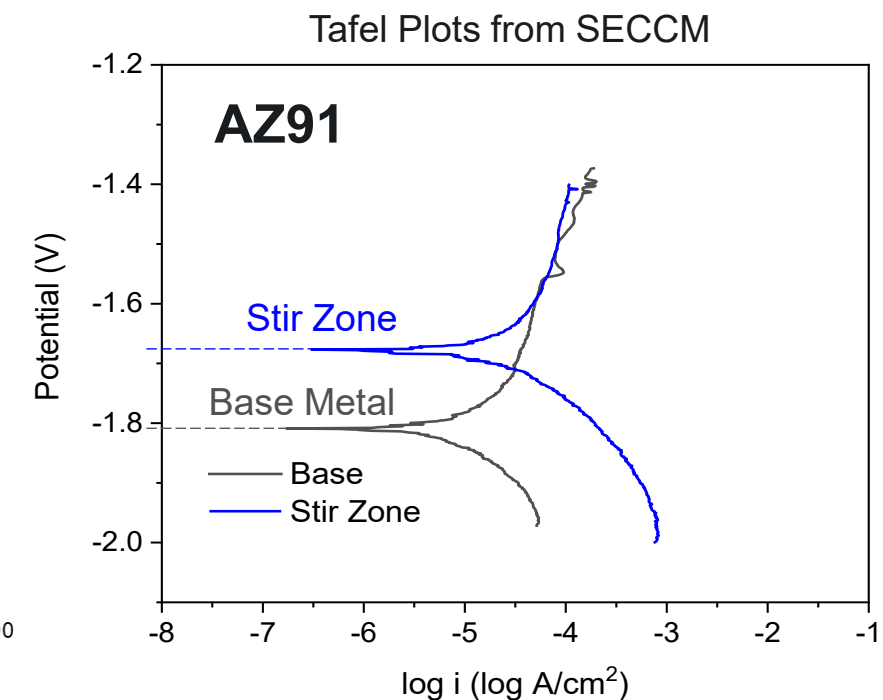
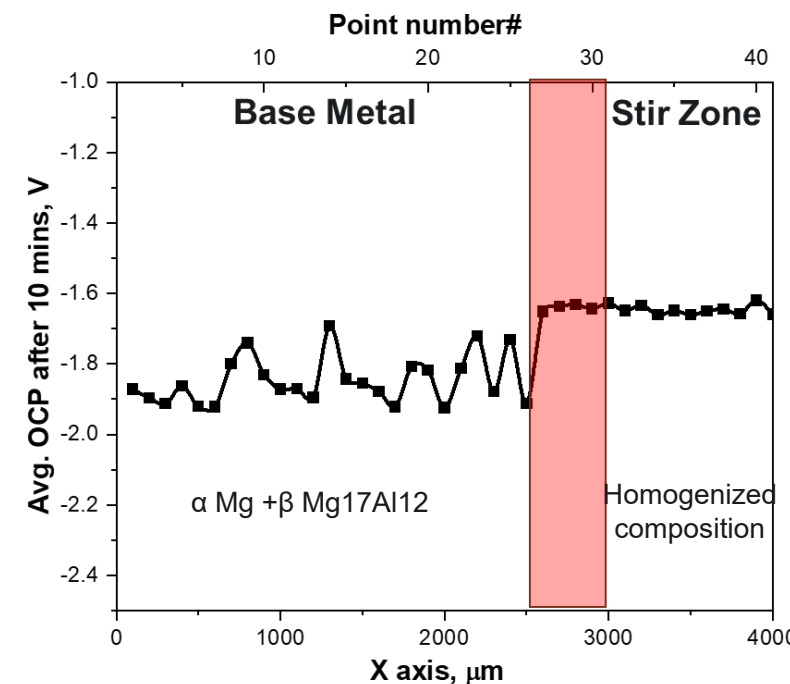
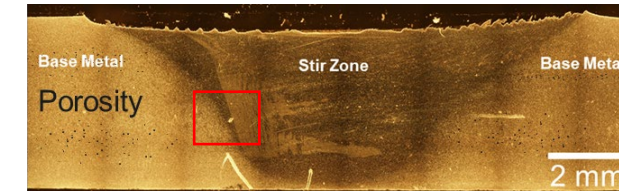
Mapping Region/Phase-Specific Corrosion Properties

Scanning Electrochemical Cell Microscopy (SECCM) Enables Localized Mapping of Corrosion Behavior

Scanning Electrochemical Cell Impedance Microscopy (SECCIM)



Implementing SECCM on FSP AZ91

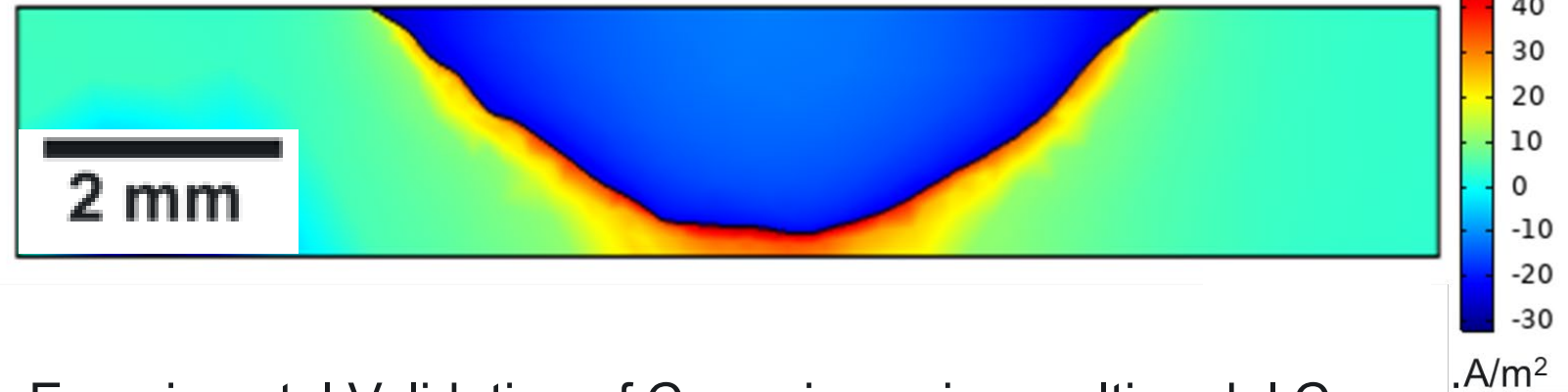


Site specific corrosion properties from SECCM can be input into COMSOL

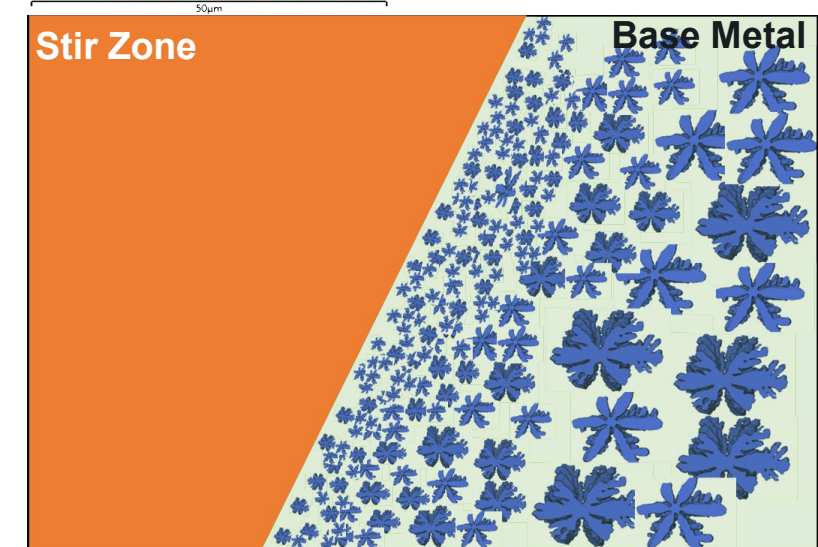
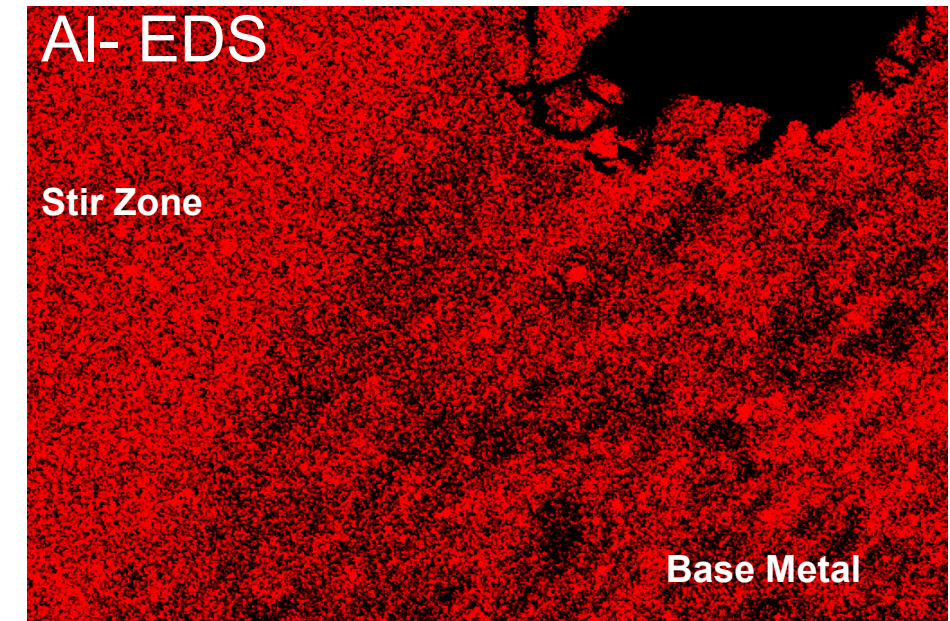
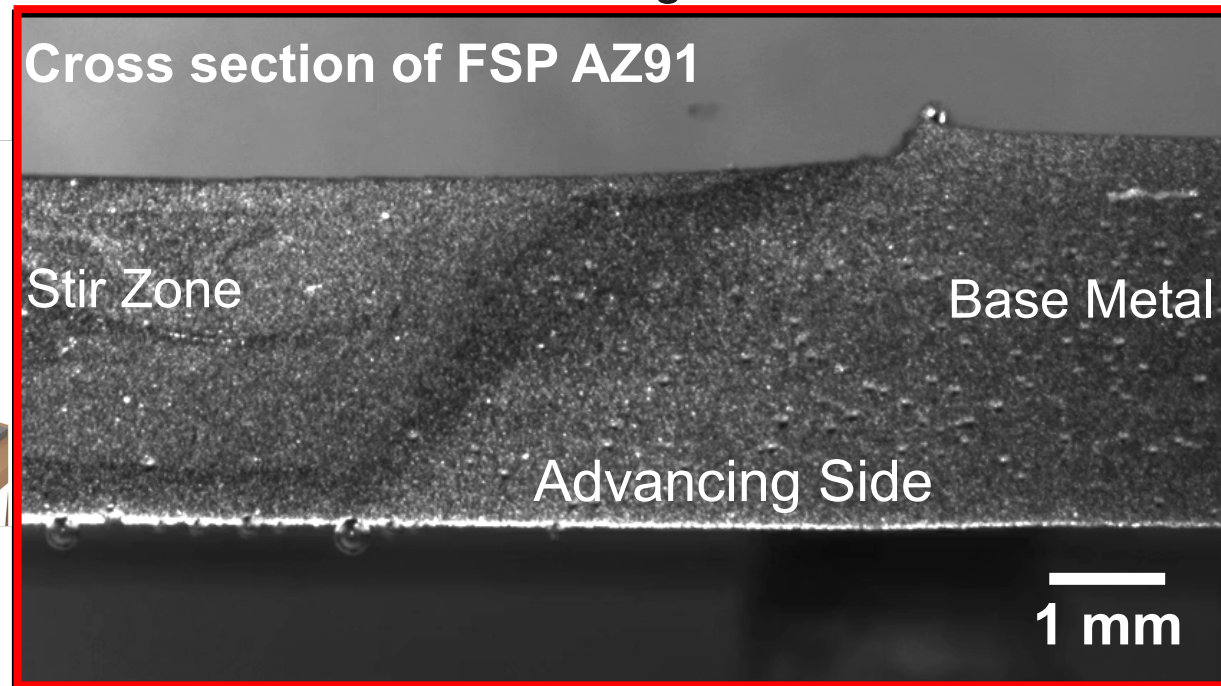
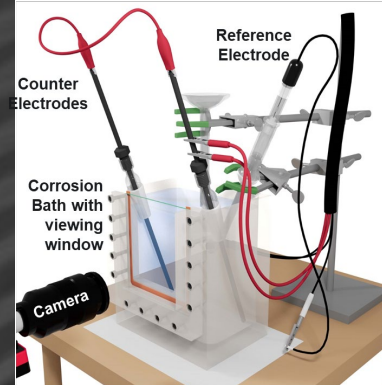
Modeling Assisted Prediction and Experimental Validation of Corrosion

COMSOL Simulations accurately predict precise locations of corrosion attack

COMSOL Modeling of Corrosion in FSP AZ91



Experimental Validation of Corrosion using multimodal Corrosion and Imaging



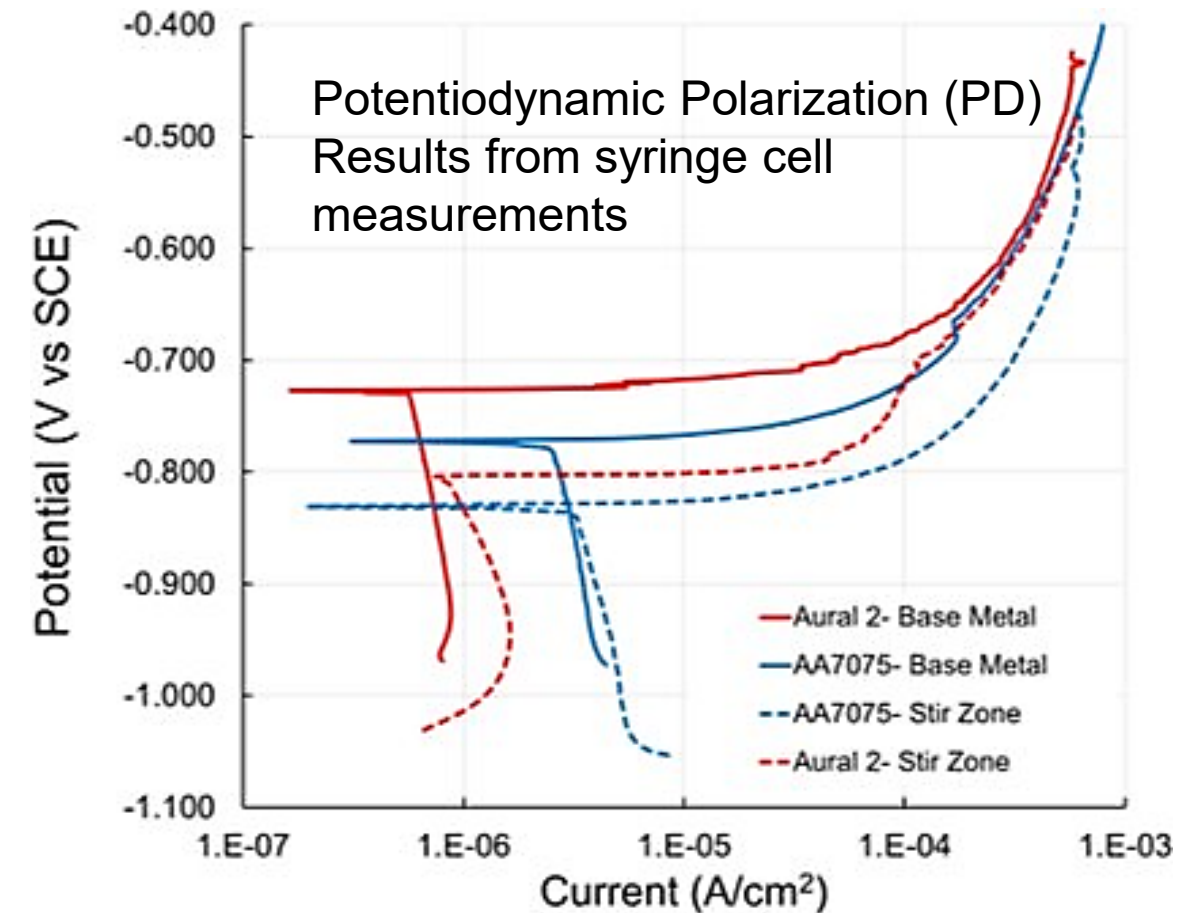
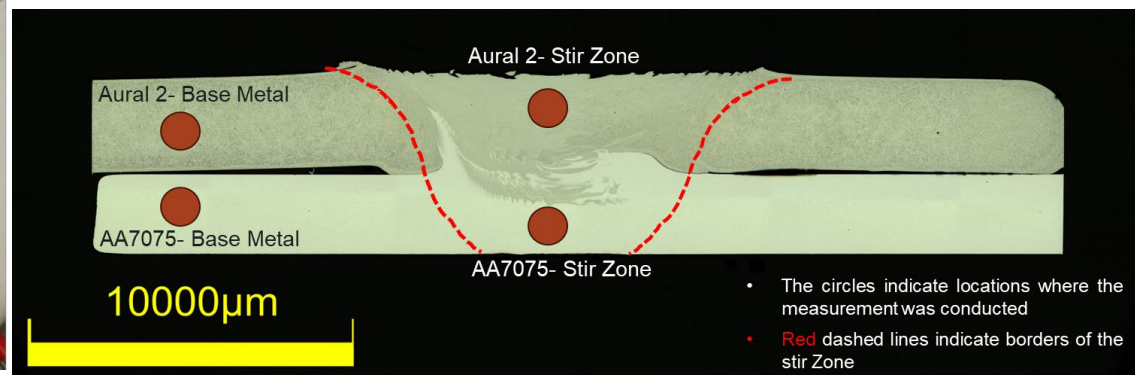
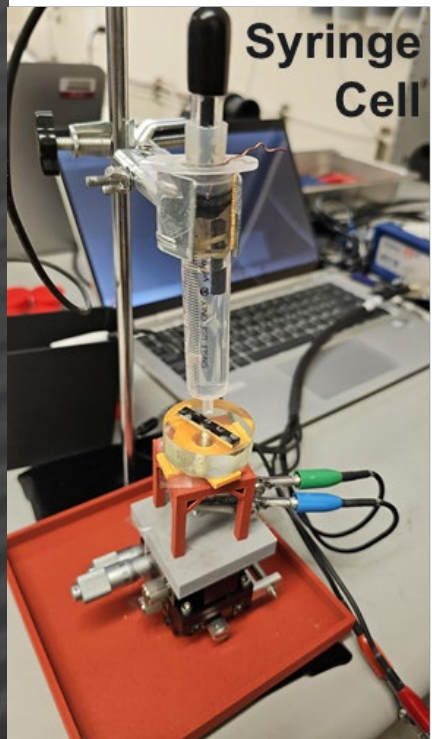
- Uniform distribution of Al
- Al rich eutectic beta phase
- Mg rich matrix



Stir Zone and Base Metal form a Galvanic Couple

Expanding Predictive Approach to FSW Joints

Using localized corrosion measurements as input to COMSOL simulation



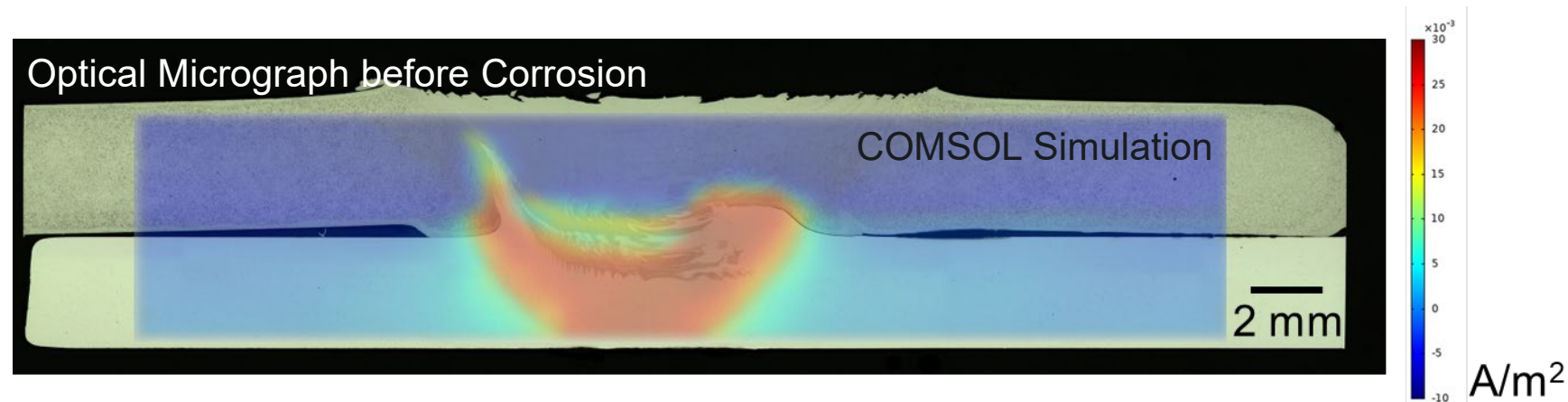
Aural2-Base Metal > AA7075 Base Metal > Aural2 Stir Zone > AA7075 Stir Zone

Each Region/Phase in the microstructure shows unique corrosion properties
Which of them is MOST SUSCEPTIBLE to corrosion?

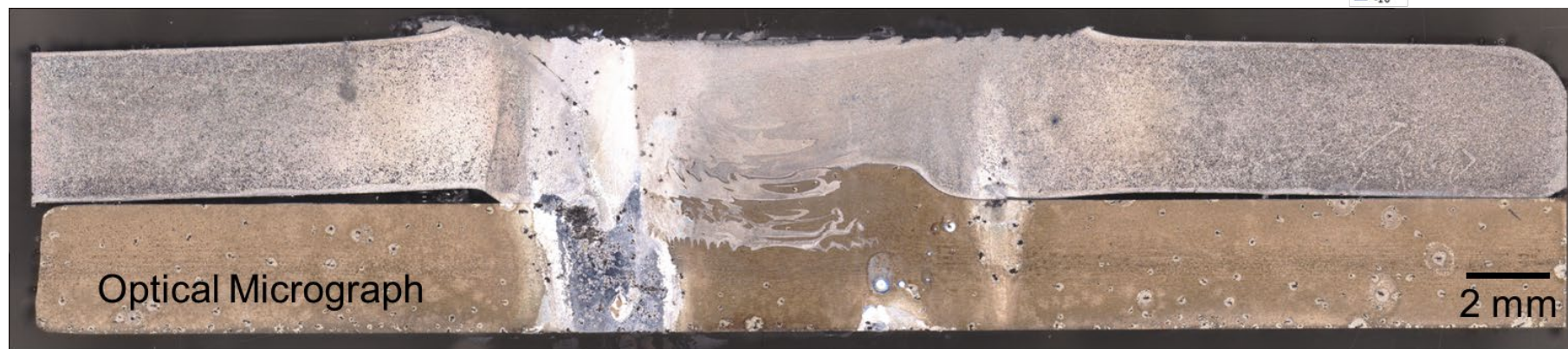
Modeling Assisted Prediction and Experimental Validation of Corrosion

Validating Simulations with Experiments

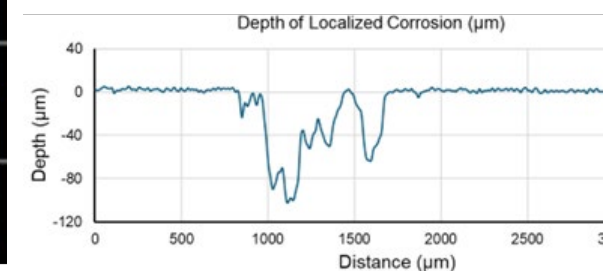
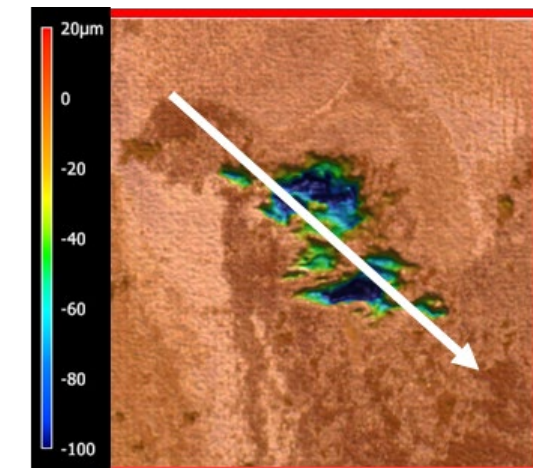
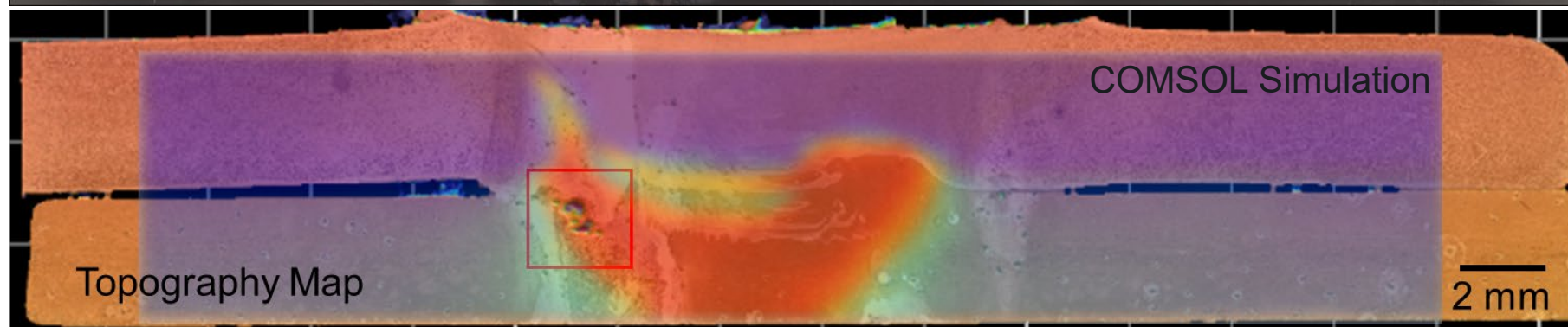
COMSOL
Simulation of
Corrosion Current
Distribution



Experimental
Validation- After
48 h of Exposure
to 3.5 wt.% NaCl
solution



Overlay of
COMSOL
simulation and
Topography map



Accurate prediction of corrosion site(s) is possible using a combination of modeling and experiments

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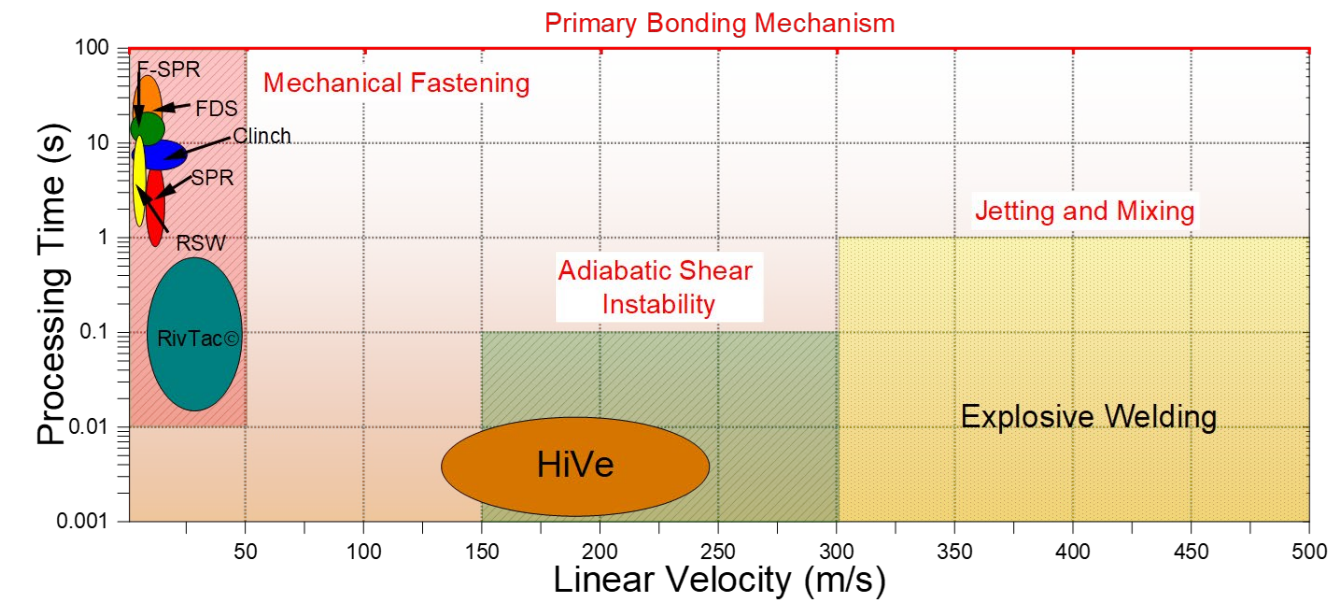
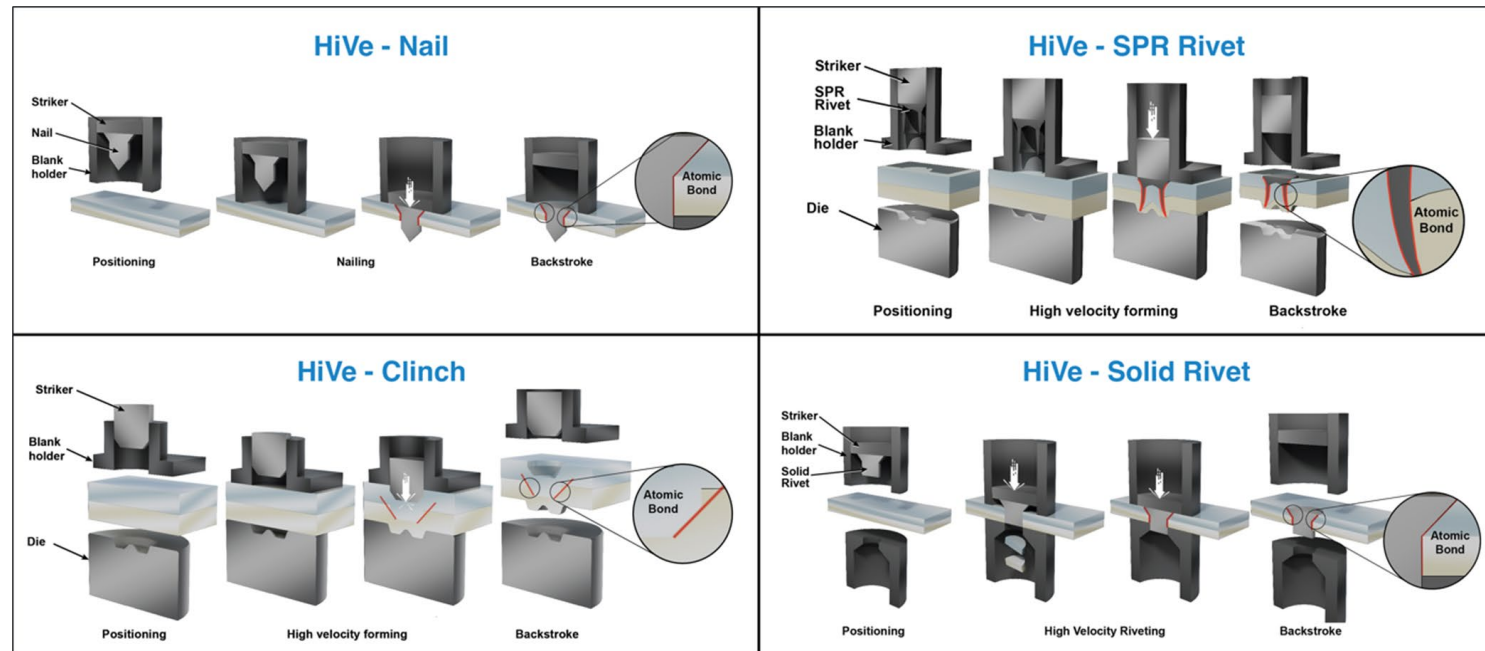
Method: Zn cold spray coating to enable barrier/sacrificial behavior

Target: Improved coating durability + longer time-to-corrosion

High Velocity (HiVe) Joining Family

Formation of Metallurgical Bond over 100 m/s

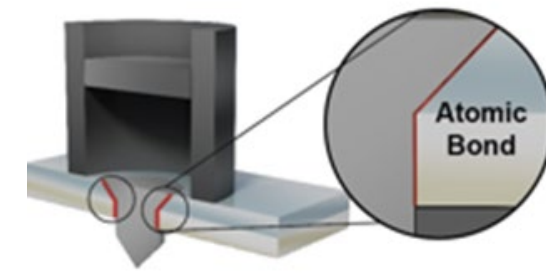
High Velocity Joining (HiVe) Methods Developed With and Without Adhesives



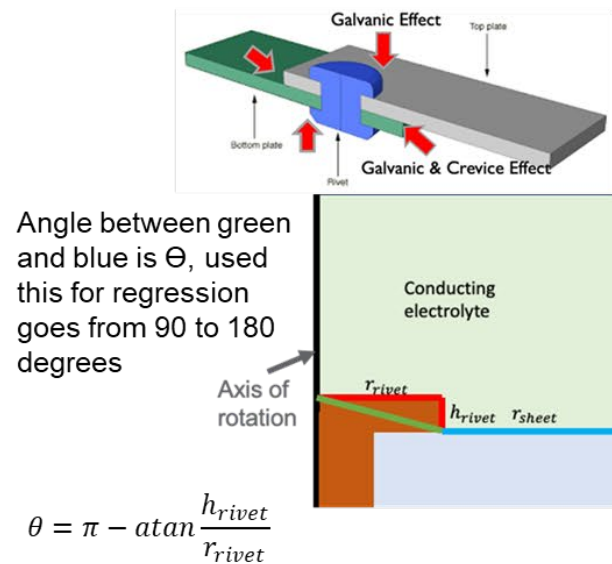
High Velocities → Locally High Impact Pressure and Temperature → **Mechanical AND Metallurgical Bonding**

Modeling of Individual HiVe Joints to Full Components

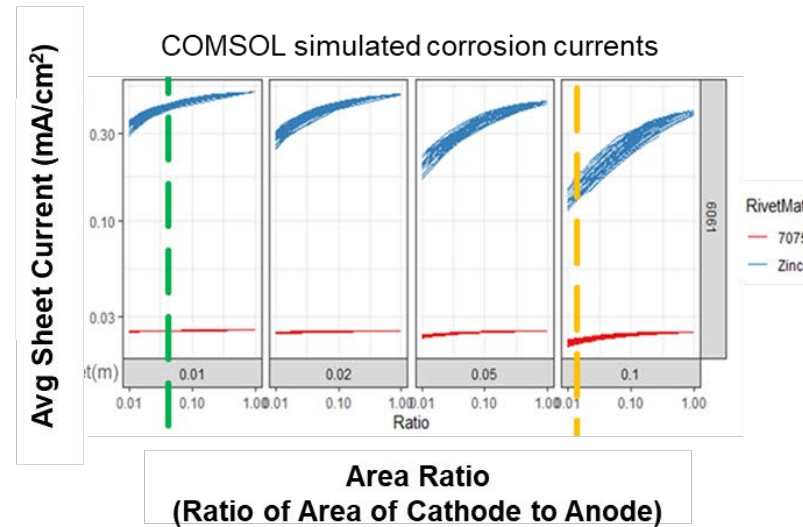
Extending joint-scale modeling of corrosion to full components



Model Setup

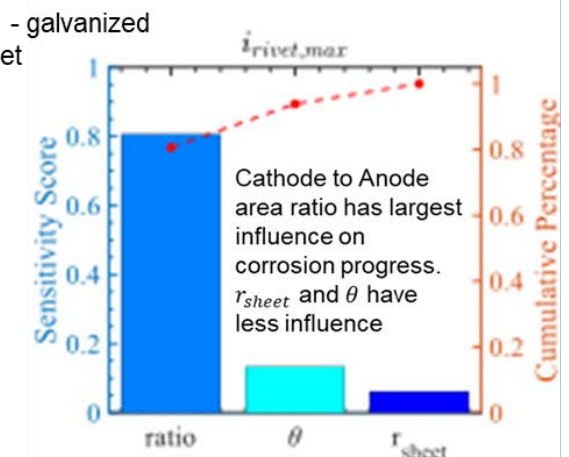


COMSOL Simulations

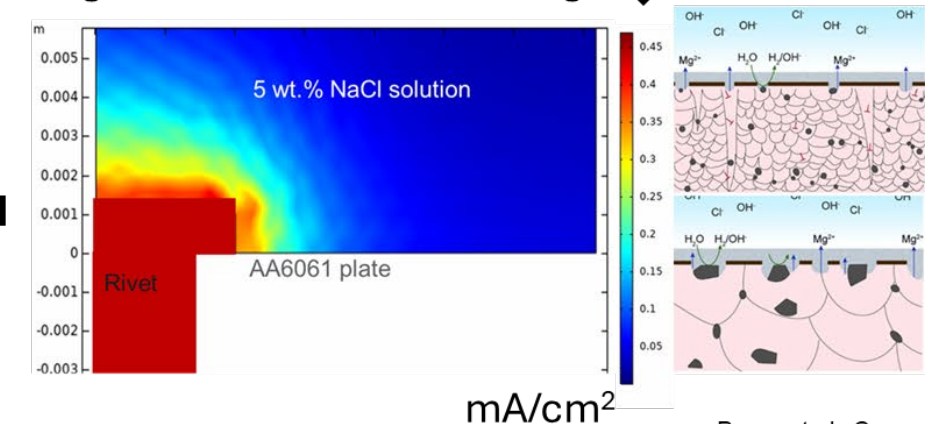


Pareto Plot of Sensitivity Score from Multivariate Adaptive Regression Splines method (MARS)

AA6061 - galvanized steel rivet

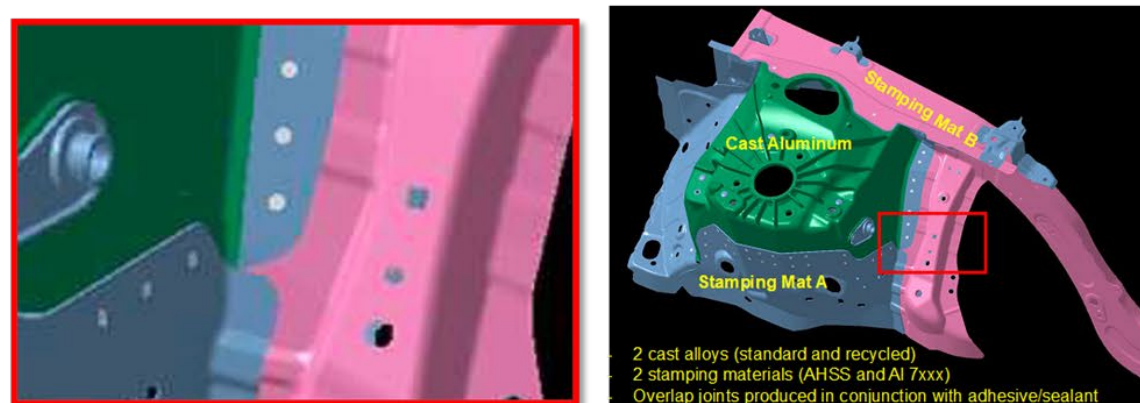


Long Term Corrosion Modeling



Beura et.al., Corrosion Science, 2022

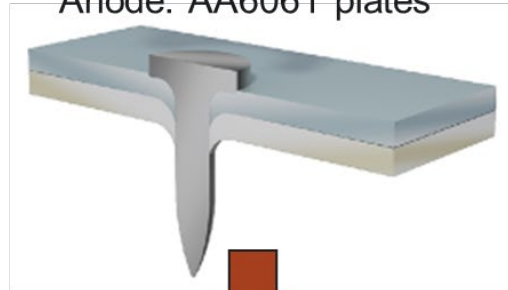
Component Level Modeling



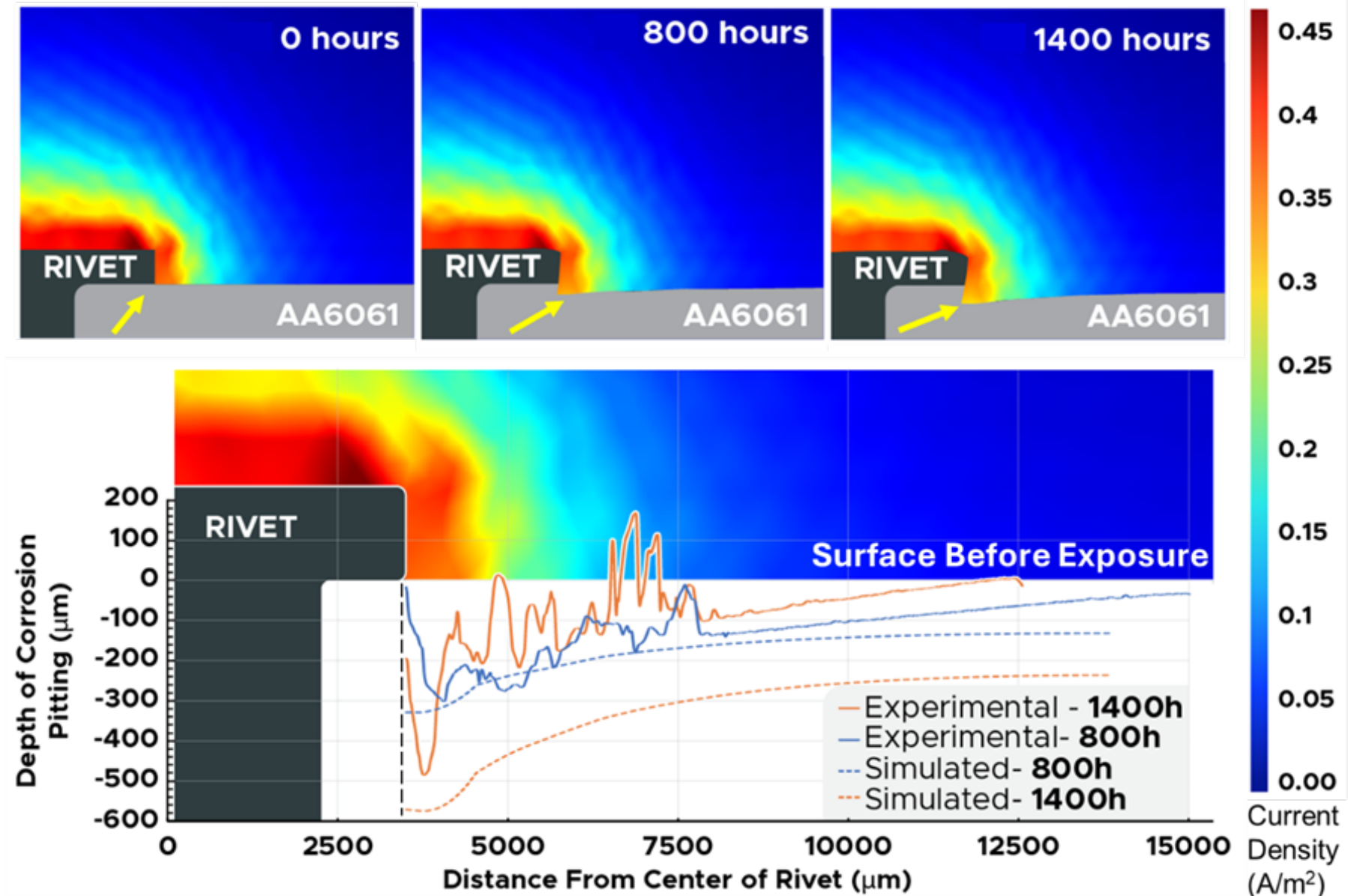
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Extending joint-scale modeling of corrosion to full components

Cathode: Galvanized steel rivet
Anode: AA6061 plates



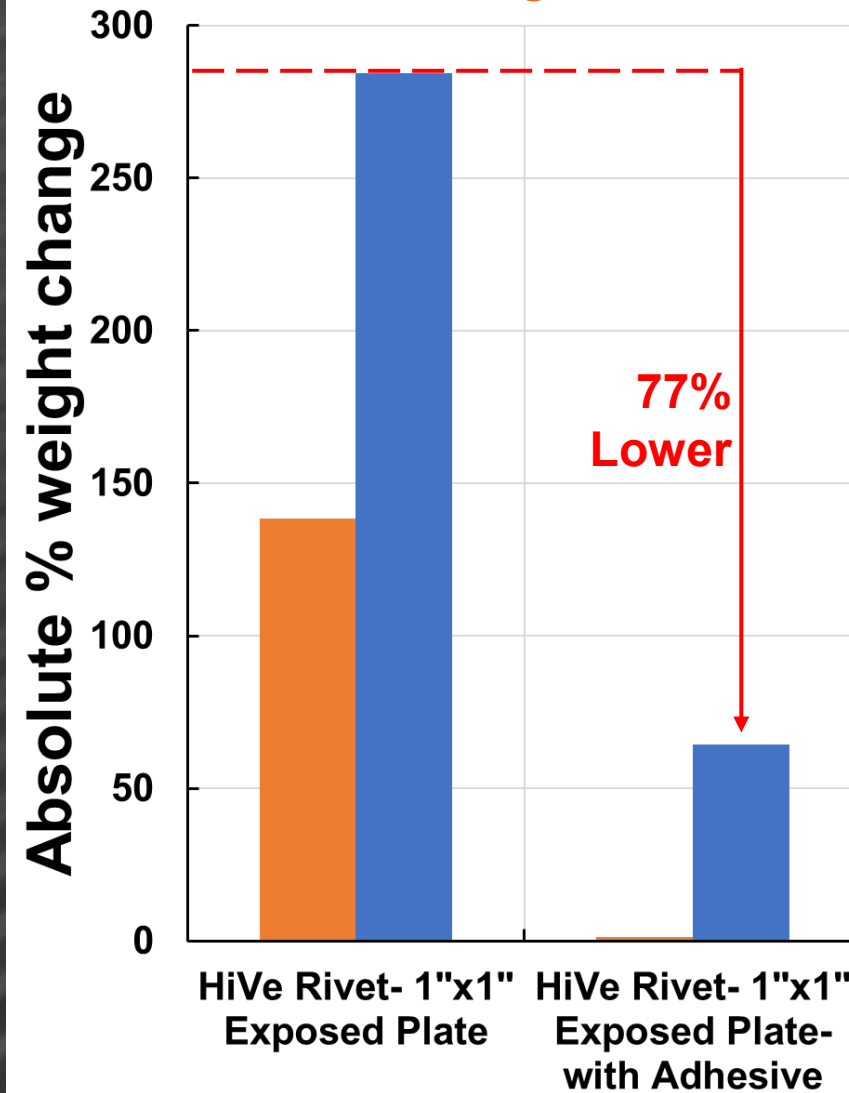
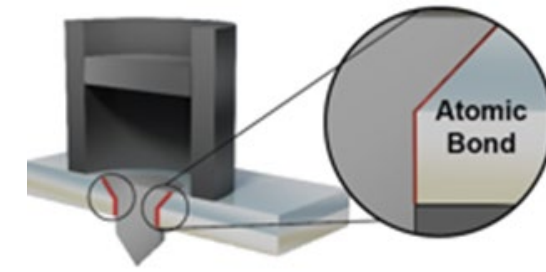
Salt fog ASTM B117 tests performed until 1400h of continuous exposure.



COMSOL simulations show concordance with maximum pit depth from experiments and can be extended to full components

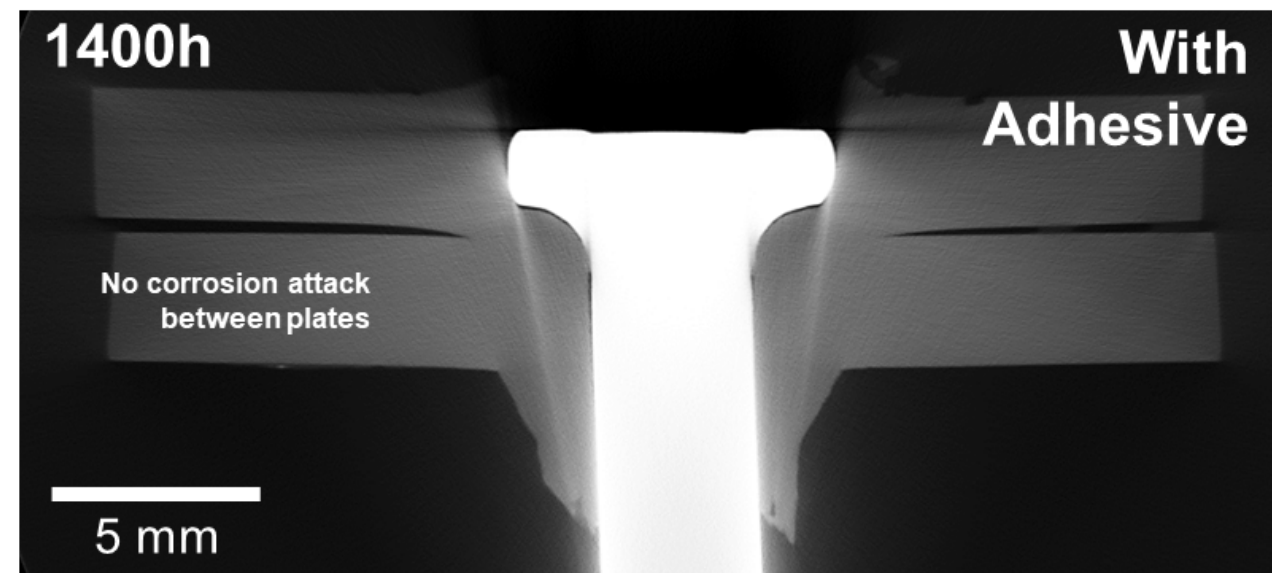
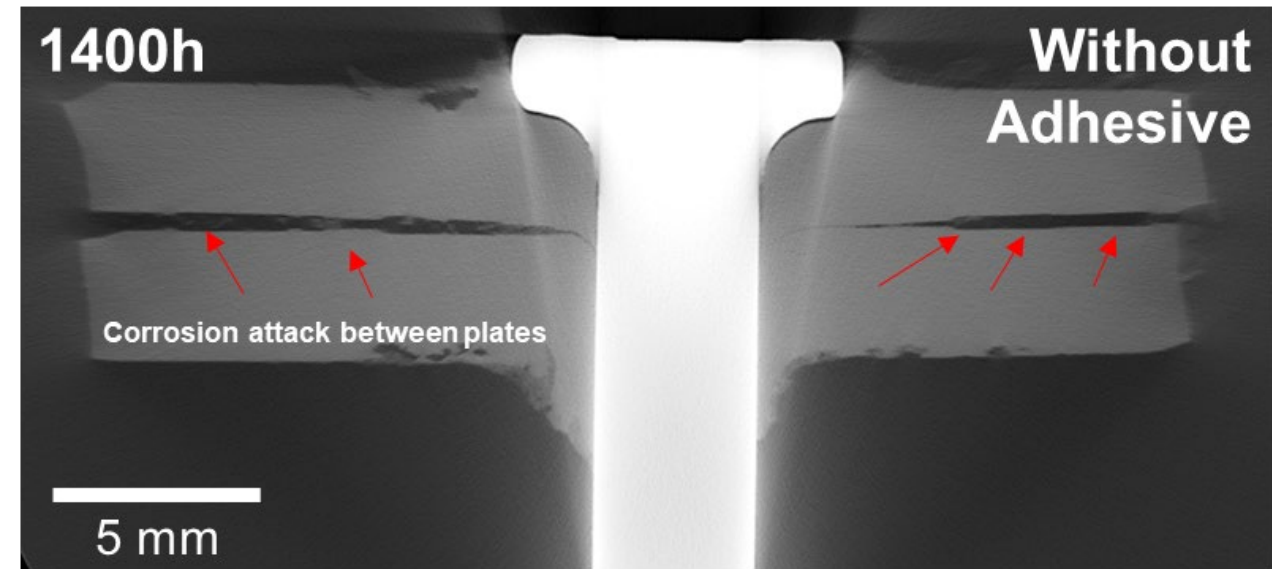
Prevent Corrosion at the Joint- Adhesives

The use of adhesives leads to lower corrosion induced weight change



Surface Preparation Conditions

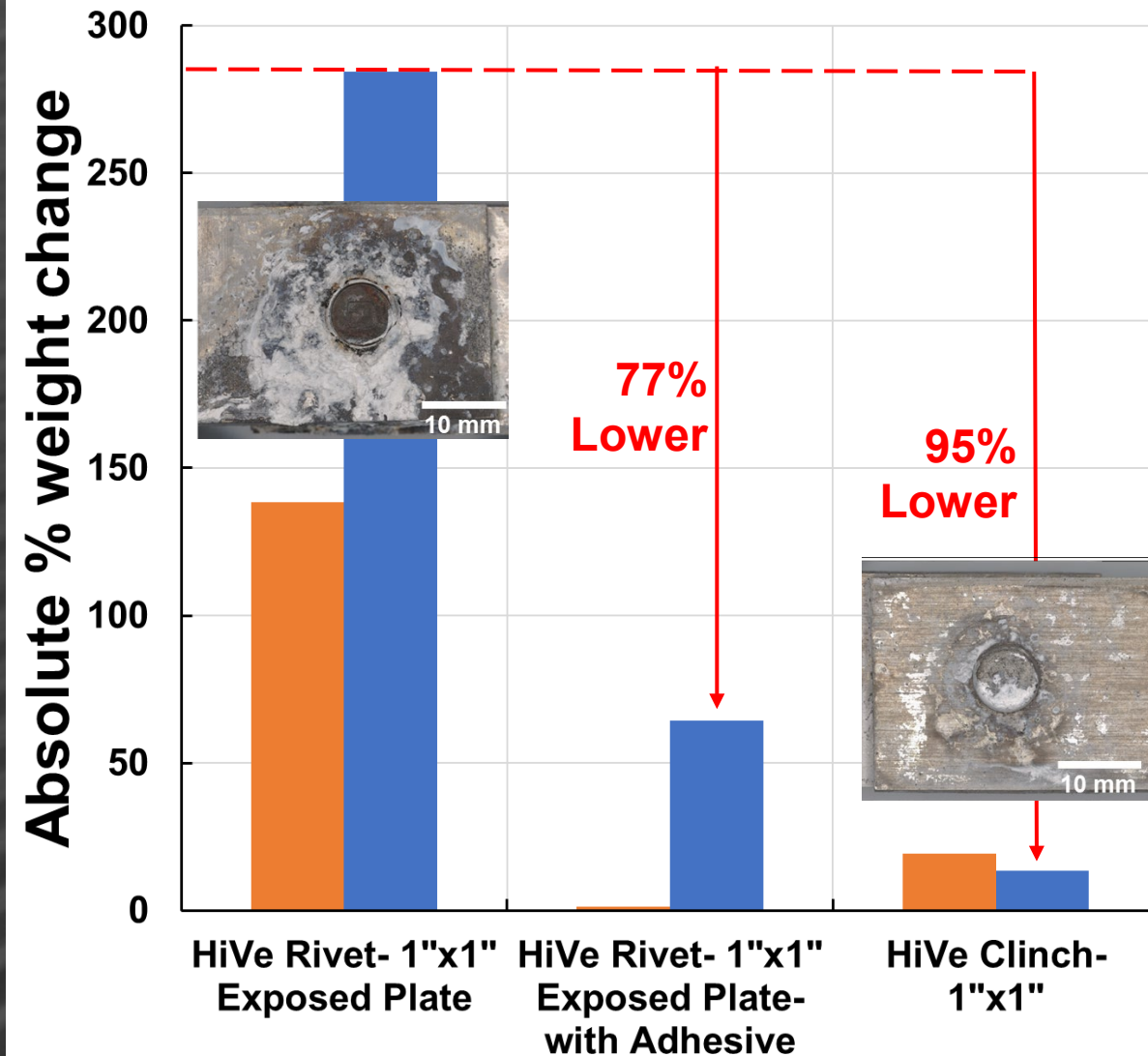
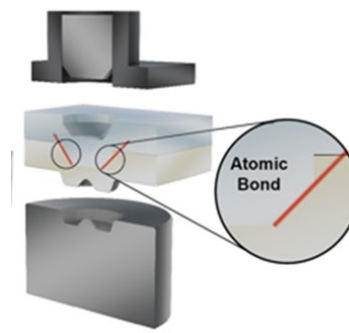
■ 800 hours
■ 1400 hours



Adhesives prevent ingress of corrosive fluid in between joints preventing crevice corrosion

Prevent Corrosion at the Joint- HiVe Clinch!

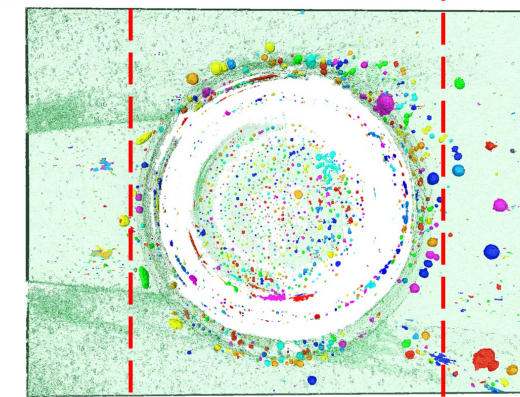
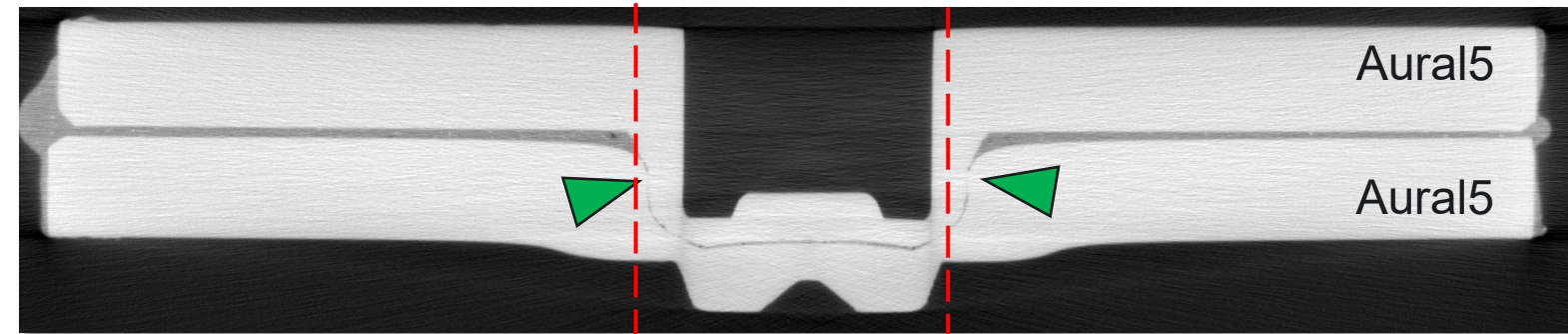
The absence of nail / rivet leads to substantially lower corrosion



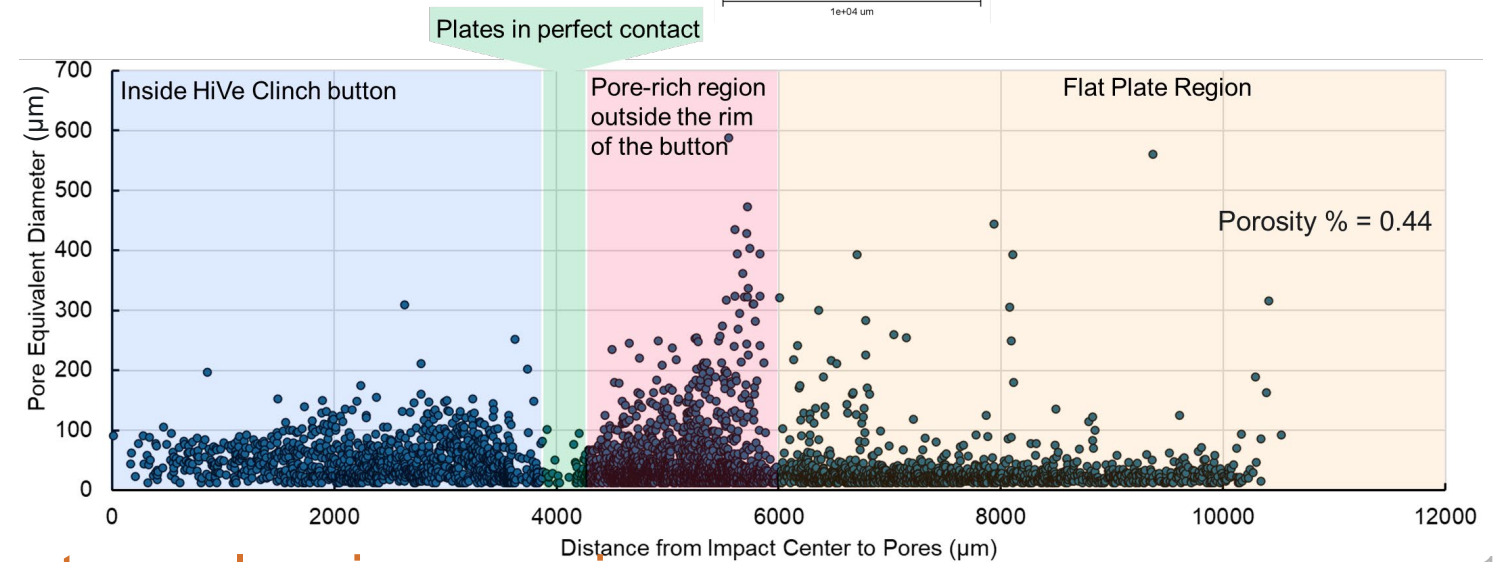
Surface Preparation Conditions

- 800 hours
- 1400 hours

HiVe Clinch mitigates galvanic corrosion



Top View of segmented XCT rendering showing adhesive and pores



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Simulate & Optimize coating → Extend lifetime

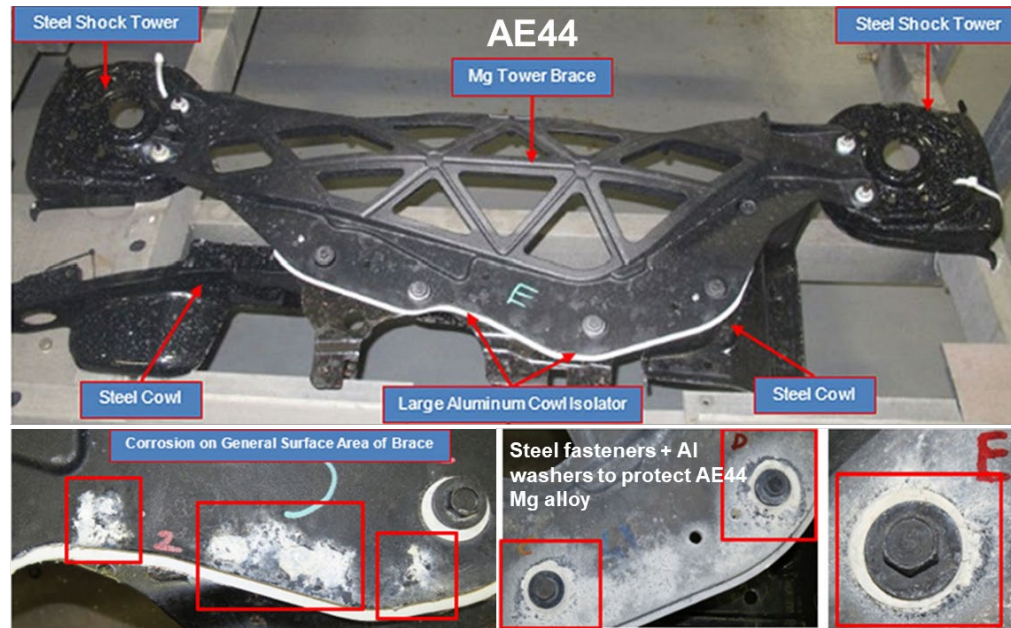
Drivers: Reactive Mg substrate + defect-sensitive coating stacks

Method: Zn cold spray coating to enable barrier/sacrificial behavior

Target: Improved coating durability + longer time-to-corrosion

The Biggest Challenge with Implementing Magnesium

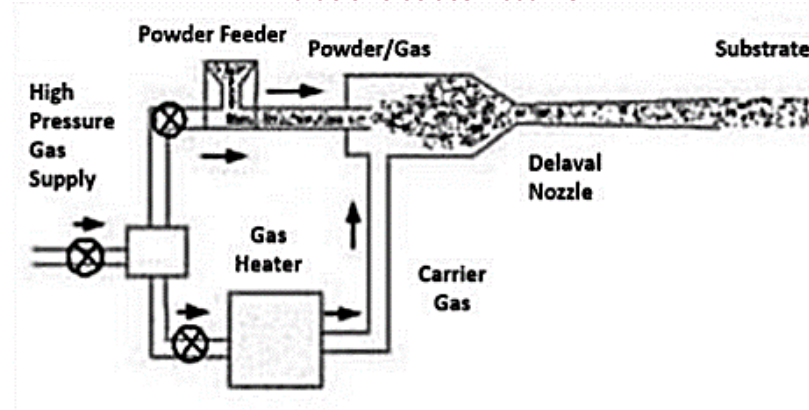
Cold sprayed coatings offer an attractive solution to localized corrosion challenges



K. Lazarz, J. Cahill, TJ Ciccone, K. Redlin, S. Simko, Corrosion Performance of a Mg tower brace, SAE technical papers, 2021

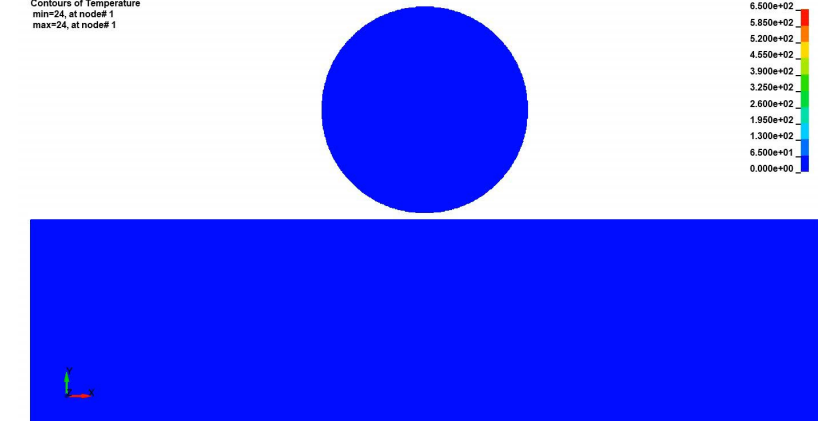
High Pressure cold spray system

Particle velocities > 600m/s



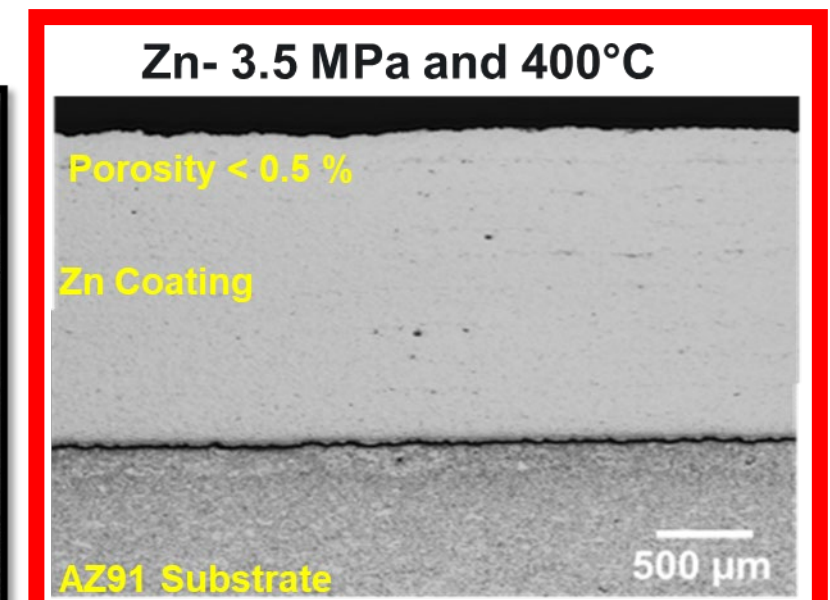
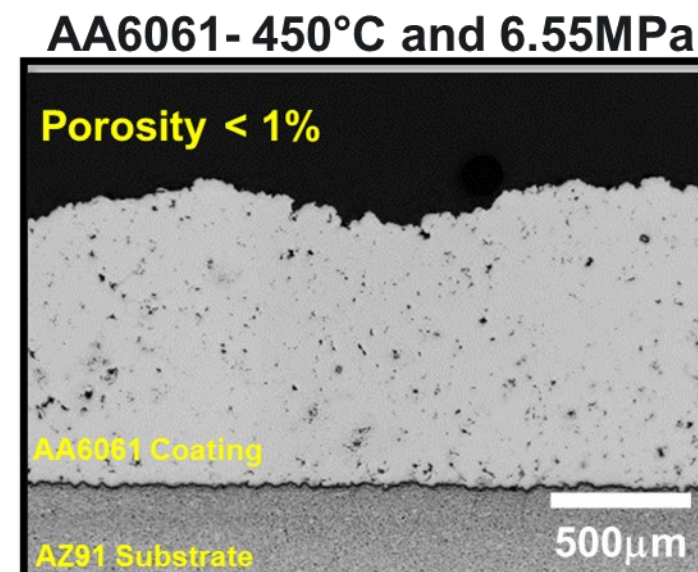
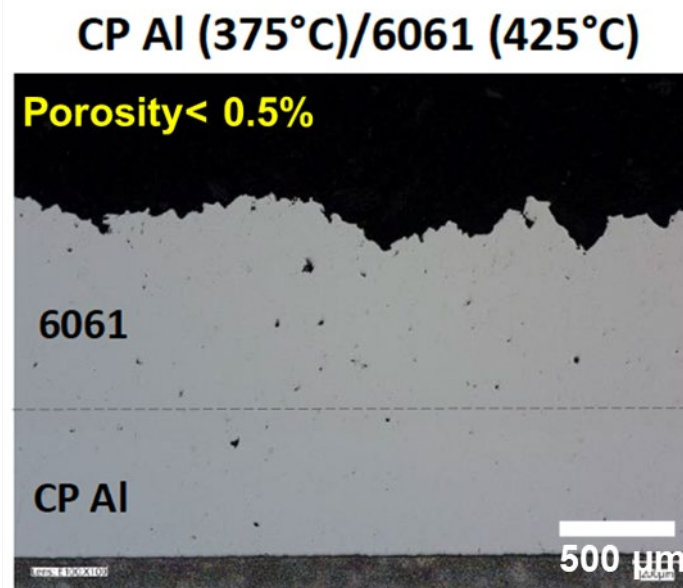
- Solid phase coating deposition.
- High deposition rate.

AZ91-1100-600ms-D26um
Time = 0
Contours of Temperature
min=24, at node# 1
max=24, at node# 1



- Metallurgical bond
- Scalable into vehicle production lines.

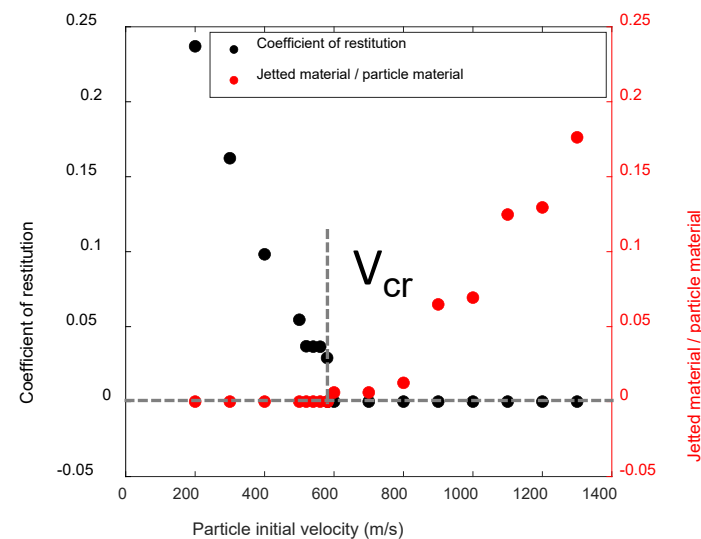
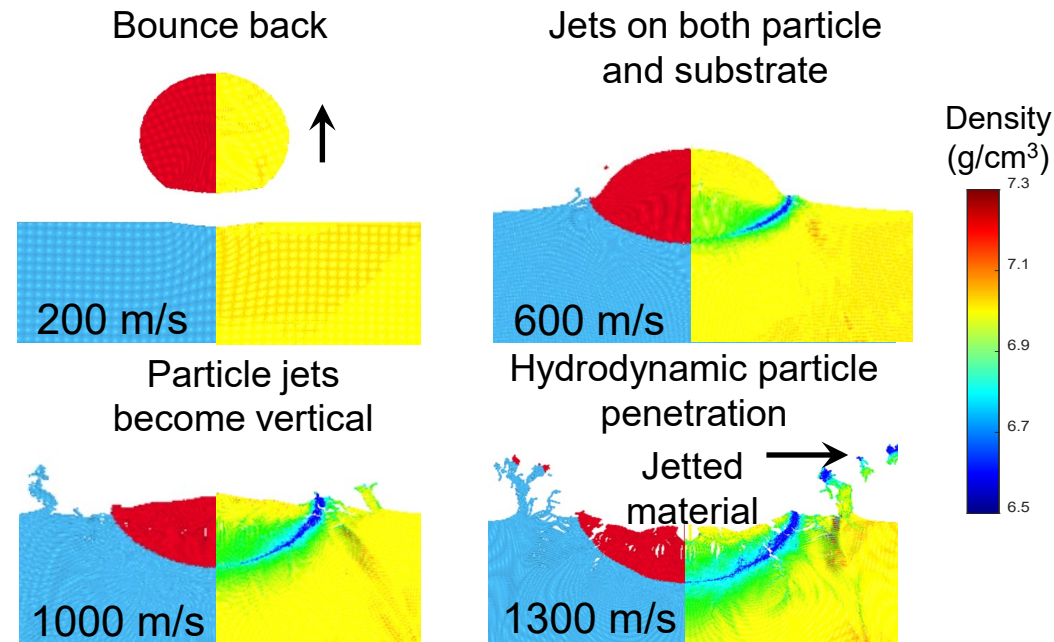
Successfully
developed and
Optimized
Cold spray
coatings



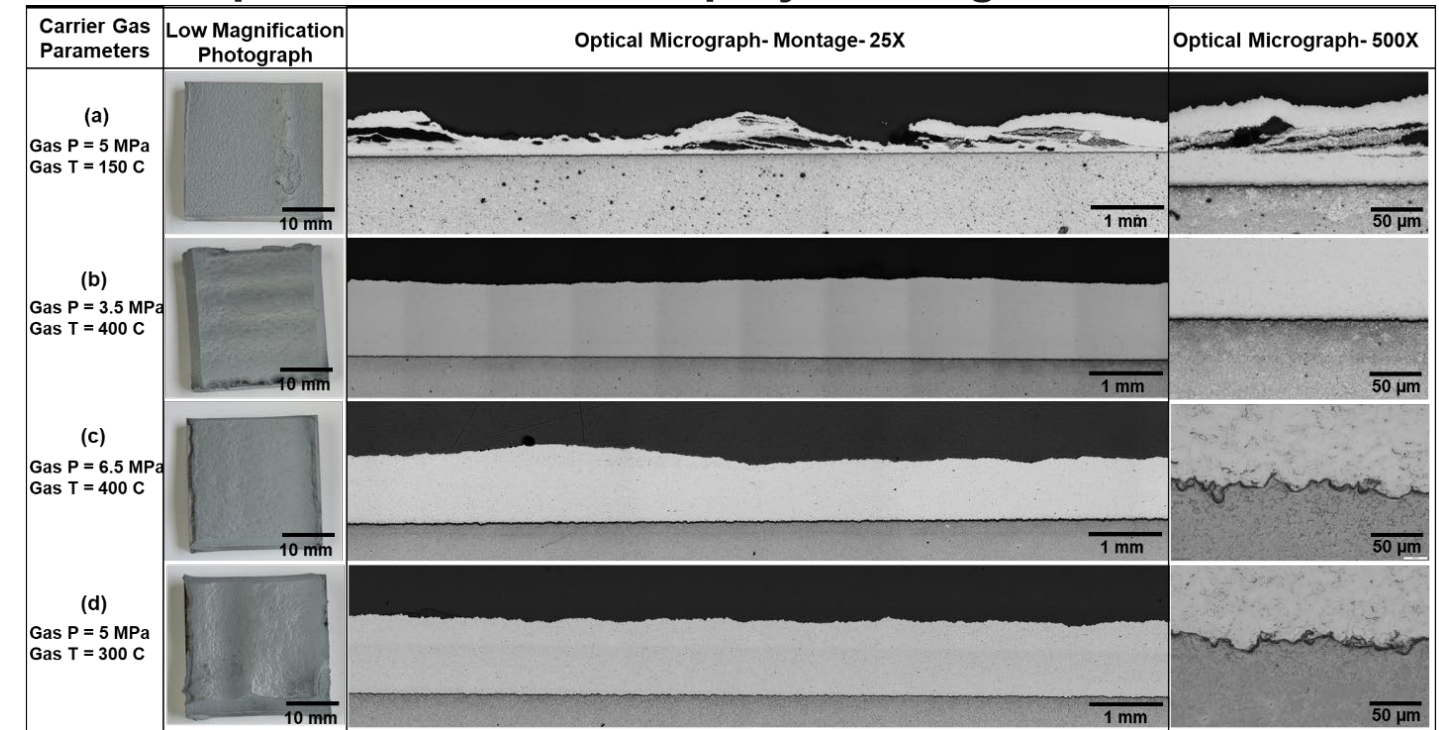
Optimization of Cold Spray Zn Coatings on AZ91

SPH can be used to predict the critical velocity for deposition

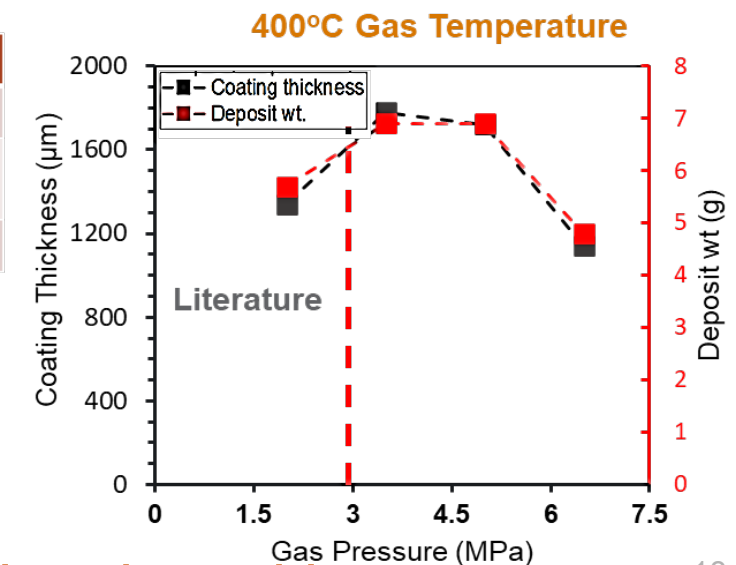
Smooth Particle Hydrodynamics to optimize the critical velocity for bonding



Optimization of Cold spray Coating Parameters



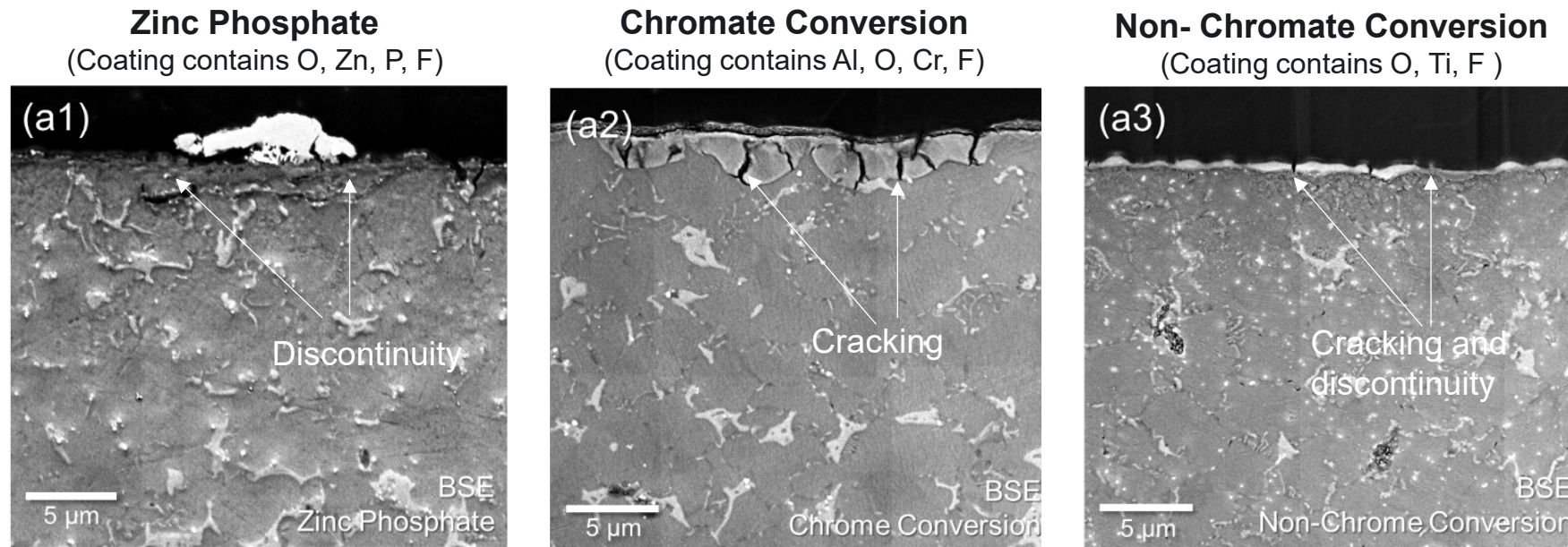
400°C Gas Temperature				
	2 MPa	3.5 MPa	5 MPa	6 MPa
Deposit thickness (mm)	1.66	1.89	1.55	1.74
Porosity (area %)	0.61	0.29	0.56	0.77



A combination of modeling and spray trials were used to optimize deposition

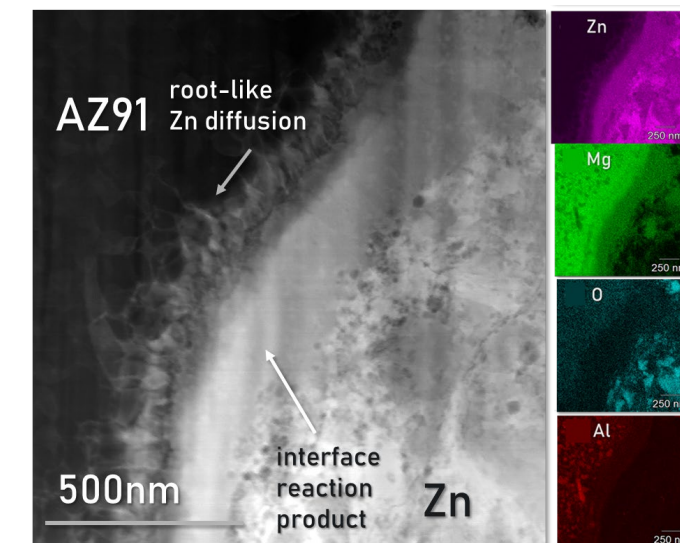
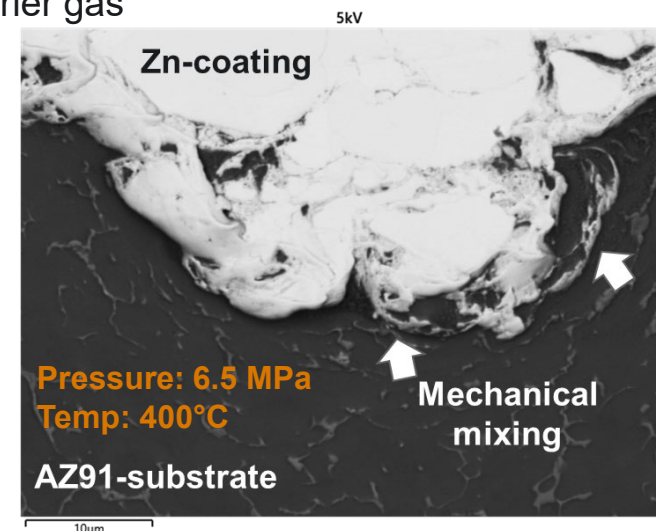
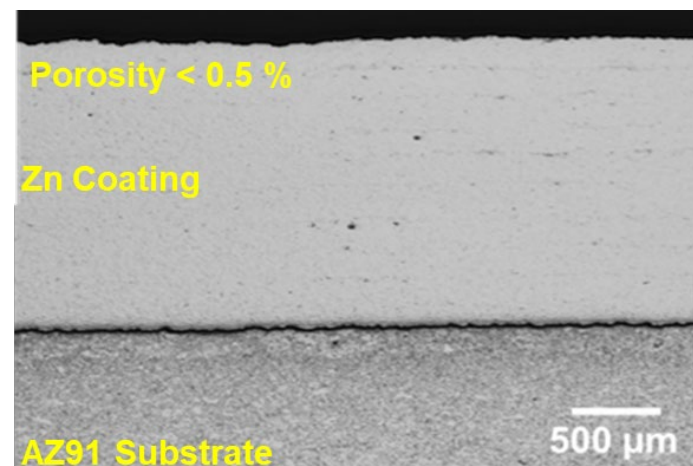
Microscopy of Pre-Treatments and Cold Sprayed Zn

Cold spray coatings of Zn offer a robust solution to the challenges associated with industrial pre-treatments



Cold Sprayed and Optimized Zn on AZ91

Optimized Zn coating- 3.5 MPa and 400°C N₂ carrier gas

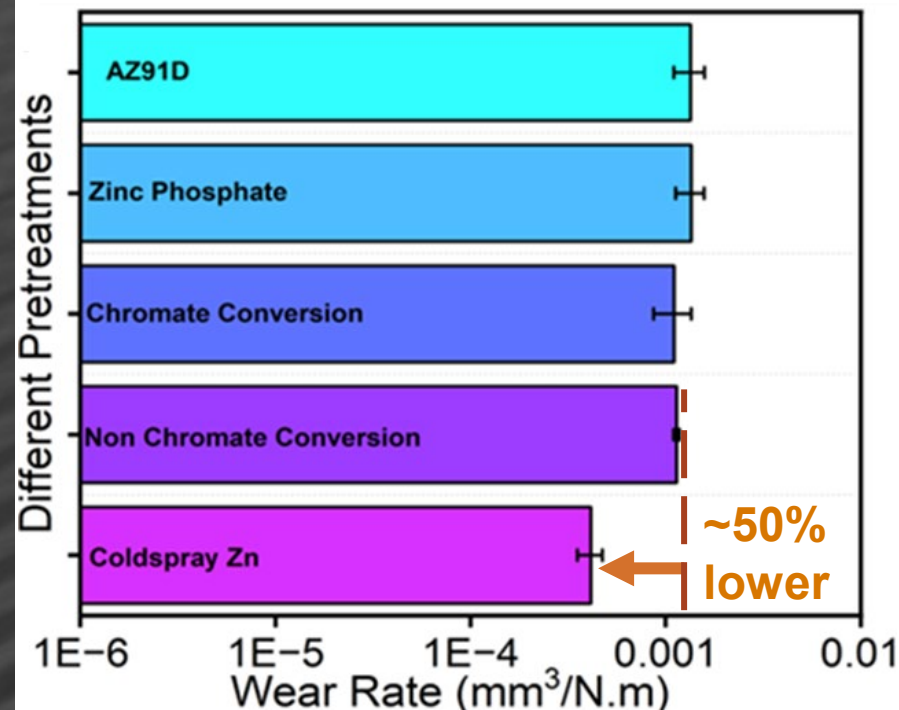


Zn coating can be selectively deposited and offers better thickness control and continuity compared to pre-treatments

Wear and Corrosion Testing of Pre-Treatments and Cold Sprayed Zn

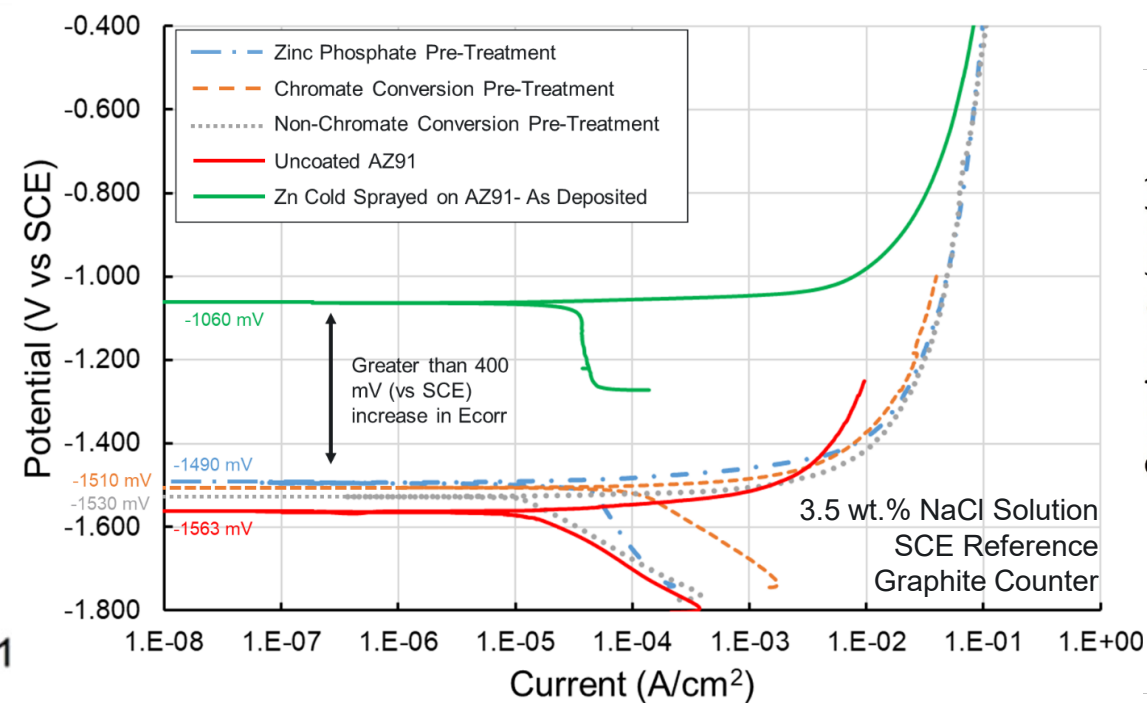
Zn cold spray coating outperforms industrial pre-treatments in terms of wear and corrosion performance

Wear Testing



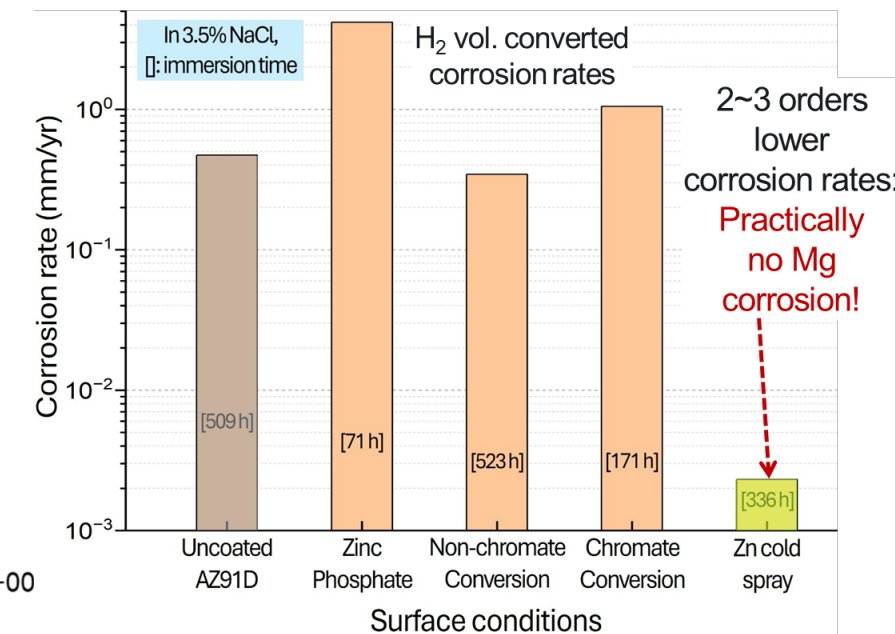
Zn cold spray coating demonstrates **50% lower wear rate** as compared to state-of-the-art pre-treatments

Potentiodynamic Polarization Testing



Zn cold spray coating demonstrates **greater than 400 mV noble E_{corr}** as compared to state-of-the-art pre-treatments

Evolved Hydrogen Collection

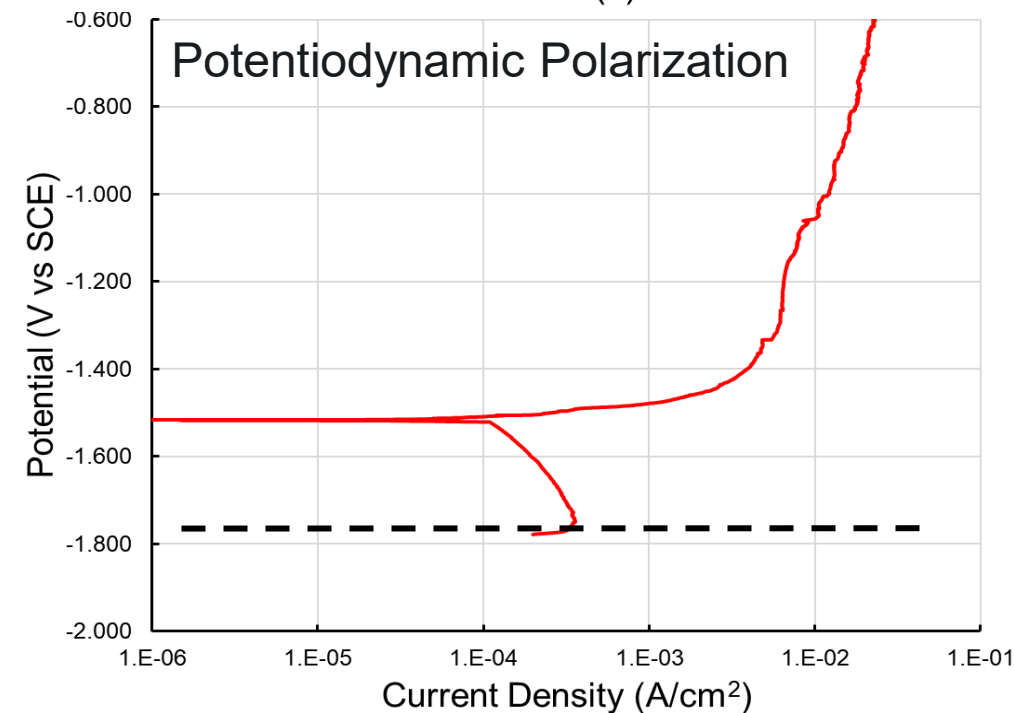
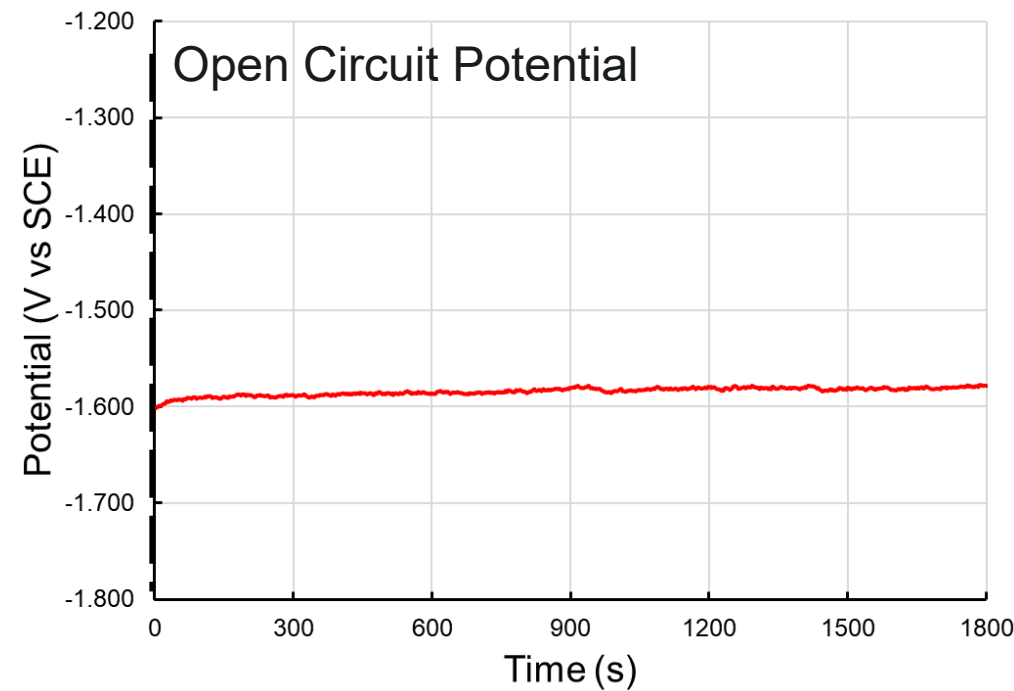


Hydrogen collection shows **negligible corrosion rate** in Zn cold spray coating

Zn cold spray coating shows lower wear rate, more positive E_{corr} and negligible corrosion rate as compared to industrial pre-treatments

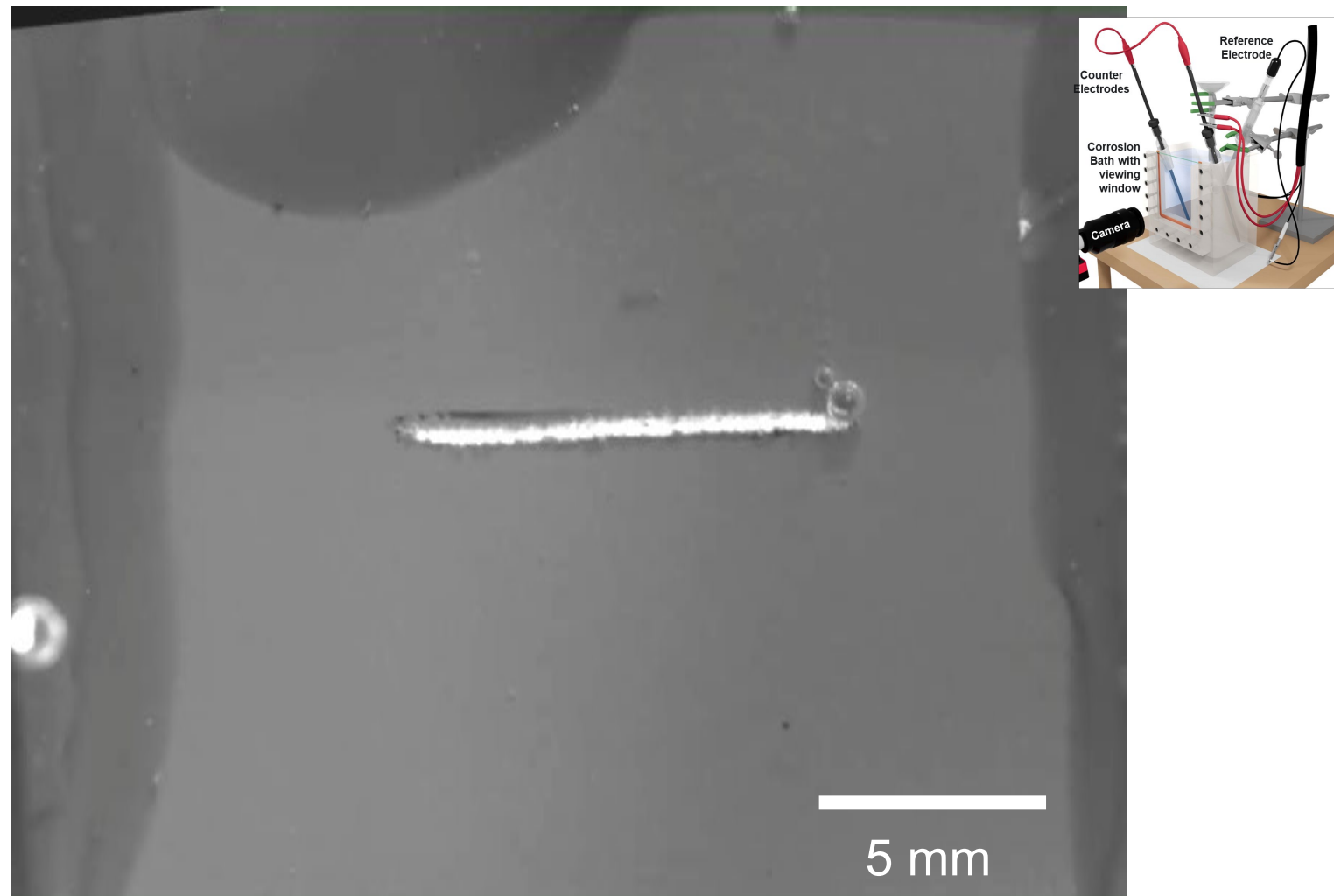
Multimodal Analysis of Accelerated Scribe Creep Corrosion

Scribe creep width indicates corrosion susceptibility and enables comparison of coatings



Substrate – AZ91
Coating- Zinc Phosphate Pre-Treatment
Ecoat + TopCoat
Scratch Length – 1 cm

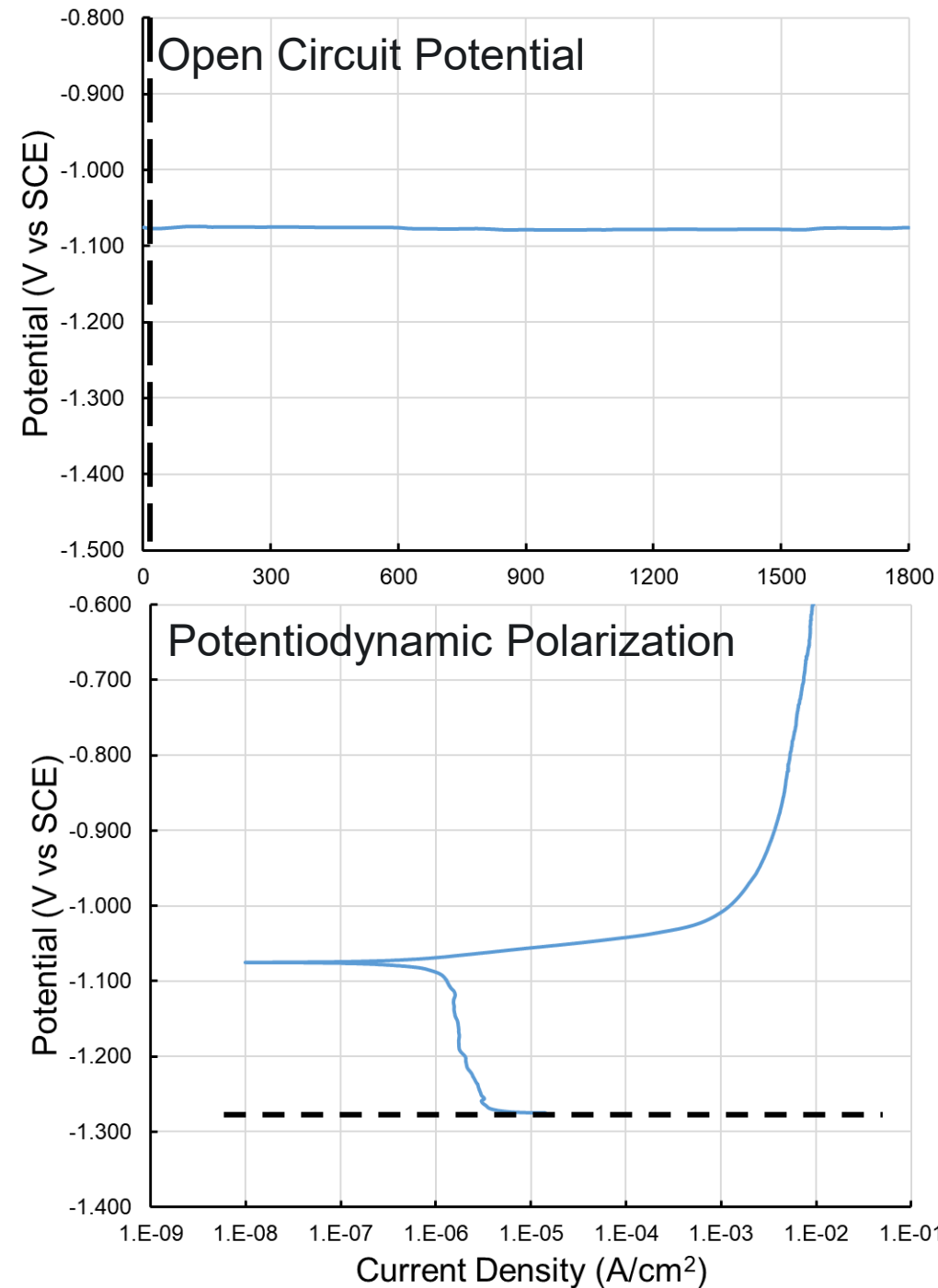
3.5 wt.% NaCl solution
SCE Reference Electrode
Graphite Counter Electrodes



Corrosion at scribe mark tears open the coating, thereby exacerbating substrate corrosion

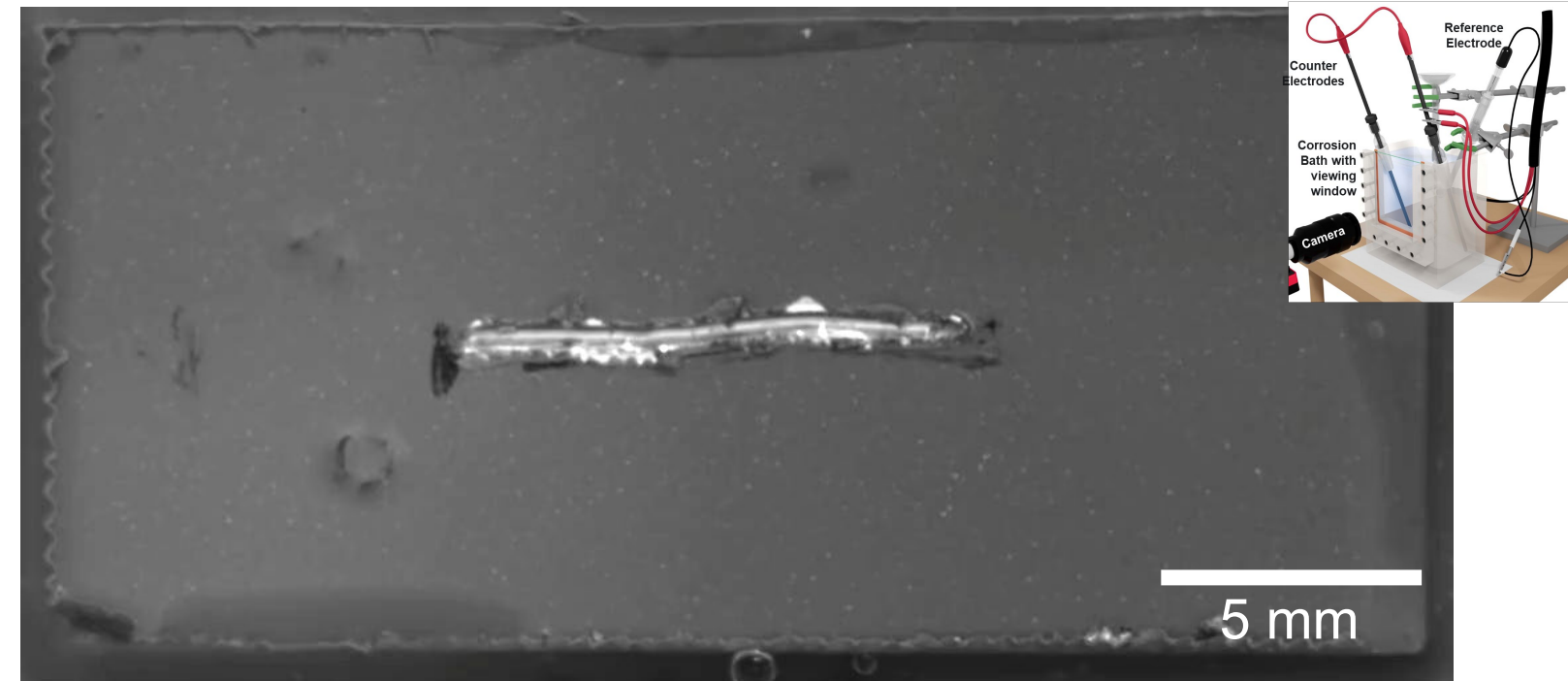
Multimodal Analysis of Accelerated Scribe Creep Corrosion

Scribe creep width indicates corrosion susceptibility and enables comparison of coatings



Substrate – AZ91
Coating- Zn cold sprayed 1 pass@45, 2 pass@90- As Deposited
Ecoat + TopCoat
Scratch Length – 1 cm

3.5 wt.% NaCl solution
SCE Reference Electrode
Graphite Counter Electrodes



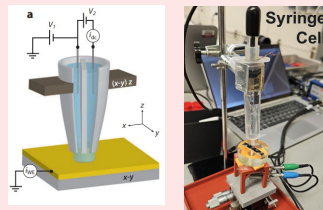
Zn cold spray coating forms a passivating oxide-hydroxide layer that stops localized corrosion

Summary

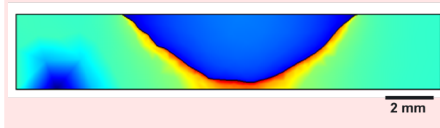
Predict

Measure locally → Model physics → Predict risk

Drivers: Electrochemistry and microstructure

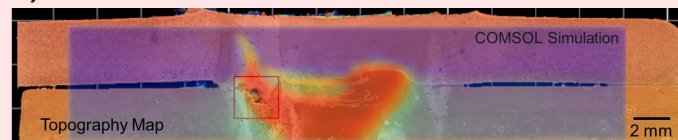


Method: SECCM / syringe cell → COMSOL



Target: Corrosion hot-spots & corrosion current map (where attack starts, how it evolves)

Overlay of COMSOL simulation and Topography map

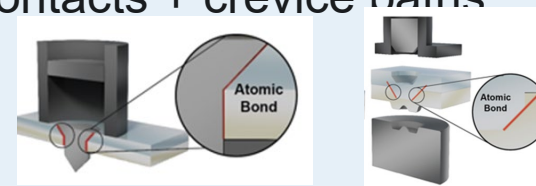


Prevent

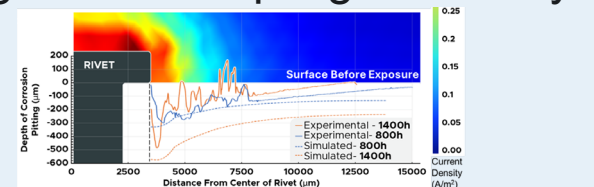
(At the Joint)

Reduce driving force → Improve longevity

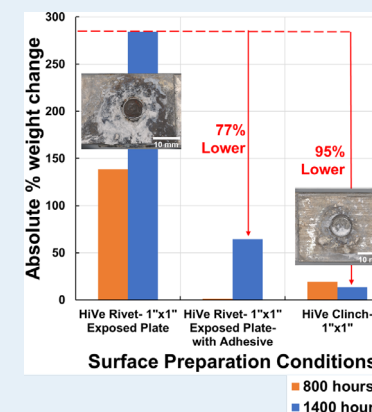
Drivers: Joint architecture + dissimilar-metal contacts + crevice paths



Method: High-velocity riveting to mitigate galvanic coupling / electrolyte access



Target: Reduced galvanic Current and delayed initiation at joints

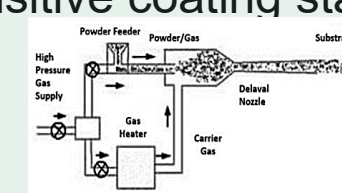


Protect

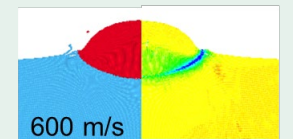
(At the Substrate)

Simulate & Optimize coating → Extend lifetime

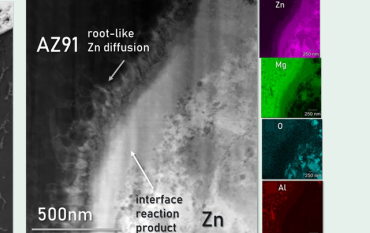
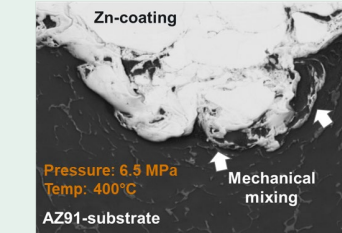
Drivers: Reactive Mg substrate + defect-sensitive coating stacks



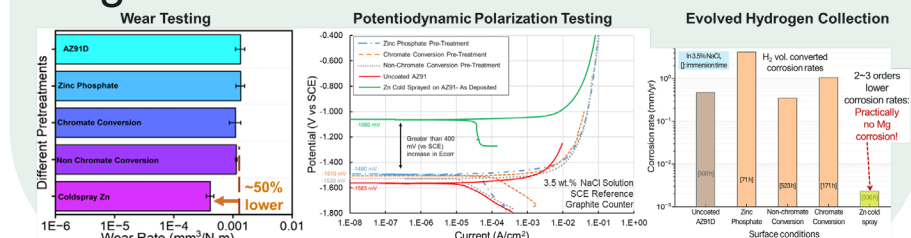
Jets on both particle and substrate



Method: Zn cold spray coating to enable barrier/sacrificial behavior



Target: Improved coating durability + longer time-to-corrosion



Thank you

1. Cold spray on cast Mg-alloys for improved corrosion properties -Invention Disclosure Submitted
2. Zn cold spray on cast and wrought Mg-alloys for improved corrosion and mechanical properties - Invention Disclosure Submitted
3. In-situ multimodal corrosion measurement system -Invention Disclosure Submitted
4. Li-salt thermal CO₂ treatment ORNL Invention Disclosure
5. Alkali metal salts facilitated CO₂ absorption induced corrosion protection layer formation on magnesium alloys“

Patents

Publications



Materials & Design
Available online 15 December 2023, 11257
In Press, Journal Pre-proof

IF = 7.9

Bond Coat Assisted Enhancement in Microstructural, Mechanical and Corrosion Behavior of AZ91 Magnesium Alloy Cold Spray Coated with Aluminum Alloys

Sridhar Niverty^{a,*}, Rajib Kalsar^a, Anthony J. Naccorelli^b, Timothy J. Eden^b, Xiaolong Ma^a, Nicole Overman^a, Glenn Grant^a, Darrell Herling^a, Vineet V. Joshi^a

scientific reports

IF = 3.4

OPEN Probing corrosion using a simple and versatile in situ multimodal corrosion measurement system

Sridhar Niverty^a, Rajib Kalsar, Robert J. Saffens, Anthony D. Gueman, Timothy J. Rosendahl, Lyndi Strange & Vineet V. Joshi^a



IF = 4.6

Lithium nitrate salt-assisted CO₂ absorption for the formation of corrosion barrier layer on AZ91D magnesium alloy[†]

Gyung G. Jang^a, Jiheon Jun^a, Jang G. Kwon^a, Yi-Feng Su^b, Mayur Pole^c, Sridhar Niverty^a and Vineet V. Joshi^a



IF = 13.8

Evaluating the bond strength and fracture mechanisms of cold-sprayed zinc coating on AZ91 magnesium substrate via a combined experimental and computational approach[†]

Tanvi Ajantiwalay^{a,*}, Lei Li^a, James V. Haag^a, Sridhar Niverty^a, Rajib Kalsar^a, Arun Devaraj^a, Ayoub Soulati^a, Vineet V. Joshi^{a,*}



IF = 13.8

Integrated process-structure–property–performance optimization of cold-sprayed zinc coatings on AZ91 magnesium alloy[†]

Sridhar Niverty^{a,*}, Rajib Kalsar^a, Lei Li^a, Nicole R. Overman^a, Jiheon Jun^a, James V. Haag^a, Tanvi A. Ajantiwalay^a, Mayur Pole^a, Vineet V. Joshi^{a,*}



IF = 5.5

Optimizing coating performance: Cold spray deposition of Al6061 powders on HPDC AZ91 alloy

Rajib Kalsar^a, Sridhar Niverty^a, Mayur Pole^a, James V. Haag^a, Tanvi Ajantiwalay^a, Vineet V. Joshi^a



IF = 2.3

Comparative Study of Zinc Cold-Spray Coating and Pre-treatments for Magnesium Alloys

SRIDHAR NIVERTY^{a,1,4}, MAYUR POLE^{a,1}, JIJO CHRISTUDASJUSTUS^{a,2}, JIHEON JUN^{a,2}, JAMES V. HAAG^{a,1}, TANVI A. AJANTI WALAY^{a,1}, RAJIB KALSAR^{a,2} and VINEET V. JOSHI^{a,1}

Invited Talks

