SAND2022-15919C

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Accelerated Corrosion Testing of Cold Spray Coatings on 304L in Chloride Environments

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Ken Ross (PNNL)

November 16, 2022

SAND2022-XXXX

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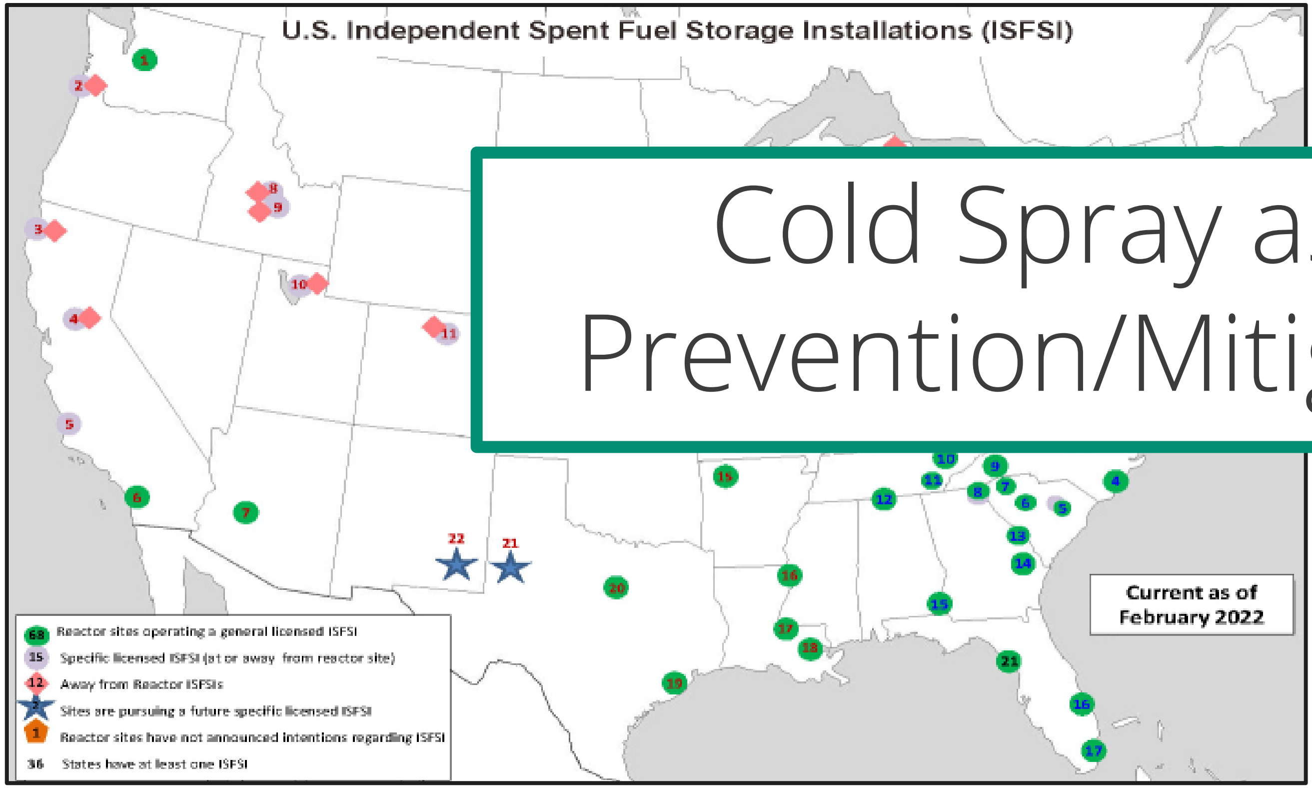
U.S. DEPARTMENT OF ENERGY

NNSA

Spent Nuclear Fuel Storage

Stainless Steel canisters- interim storage for spent nuclear fuel waste
Marine/near marine – chloride brines



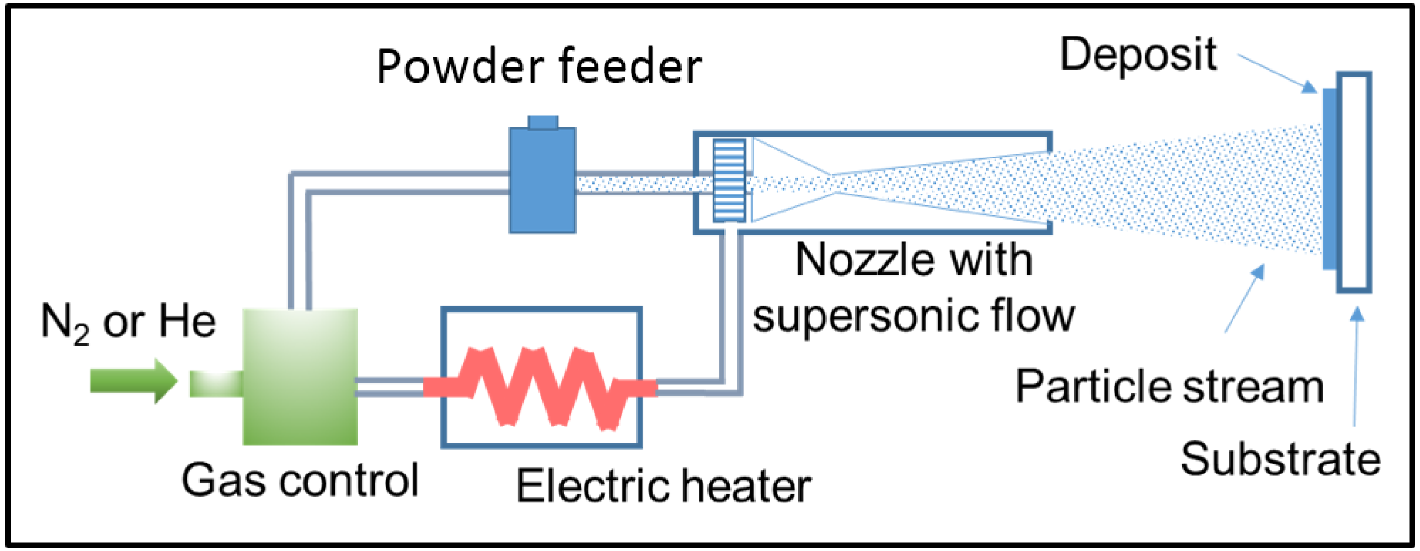
https://www.nrc.gov/reading-rm/doc-collections/maps/isfsi.html

Cold Spray as Corrosion Prevention/Mitigation Coating

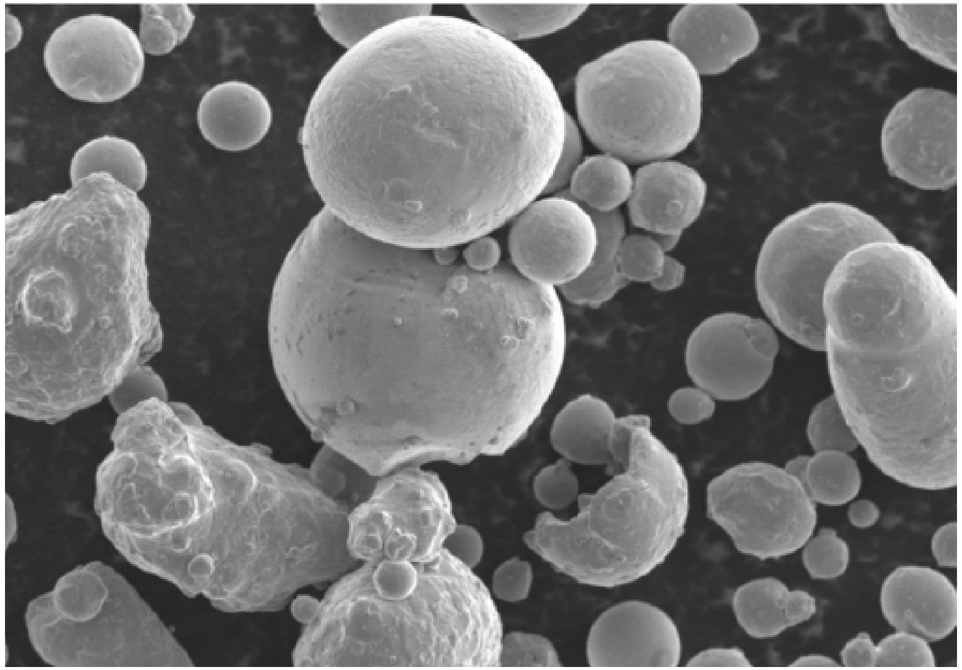
Vertical canisters in concrete overpacks
Bryan & Enos, SAND2014-20347O

Cold Spray Technique

Metal particles accelerated by a stream of inert gas into a substrate.
Adhered by plastic deformation.



High-pressure cold spray process.
Image from Ken Ross PNNL-30299. 2021.



316L stainless steel powder

Lower temperature helps prevent:

- Temperature-dependent crystalline transformations
- Oxidation
- Vaporization

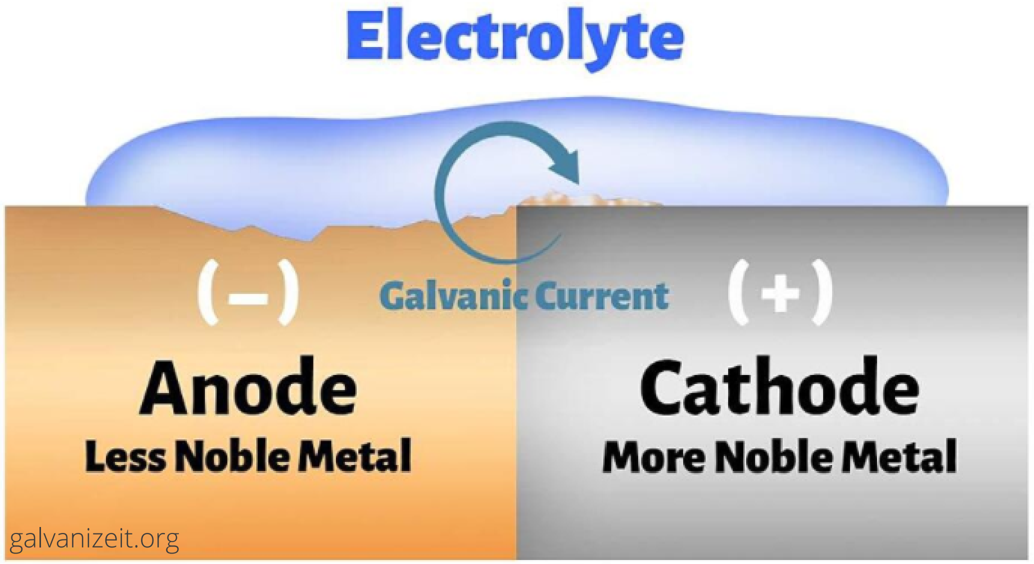
Patch Application

In-Service Canisters:
>3,000 SNF Canisters
>70 ISFSI sites

Patch Application:
focus on vulnerable/damaged areas
<u>exposed junction between substrate & cold spray</u>

Concerns at Patch Edge:

- Galvanic Corrosion
- Possible Deformation Layer
- Residual Stresses



Material Selection

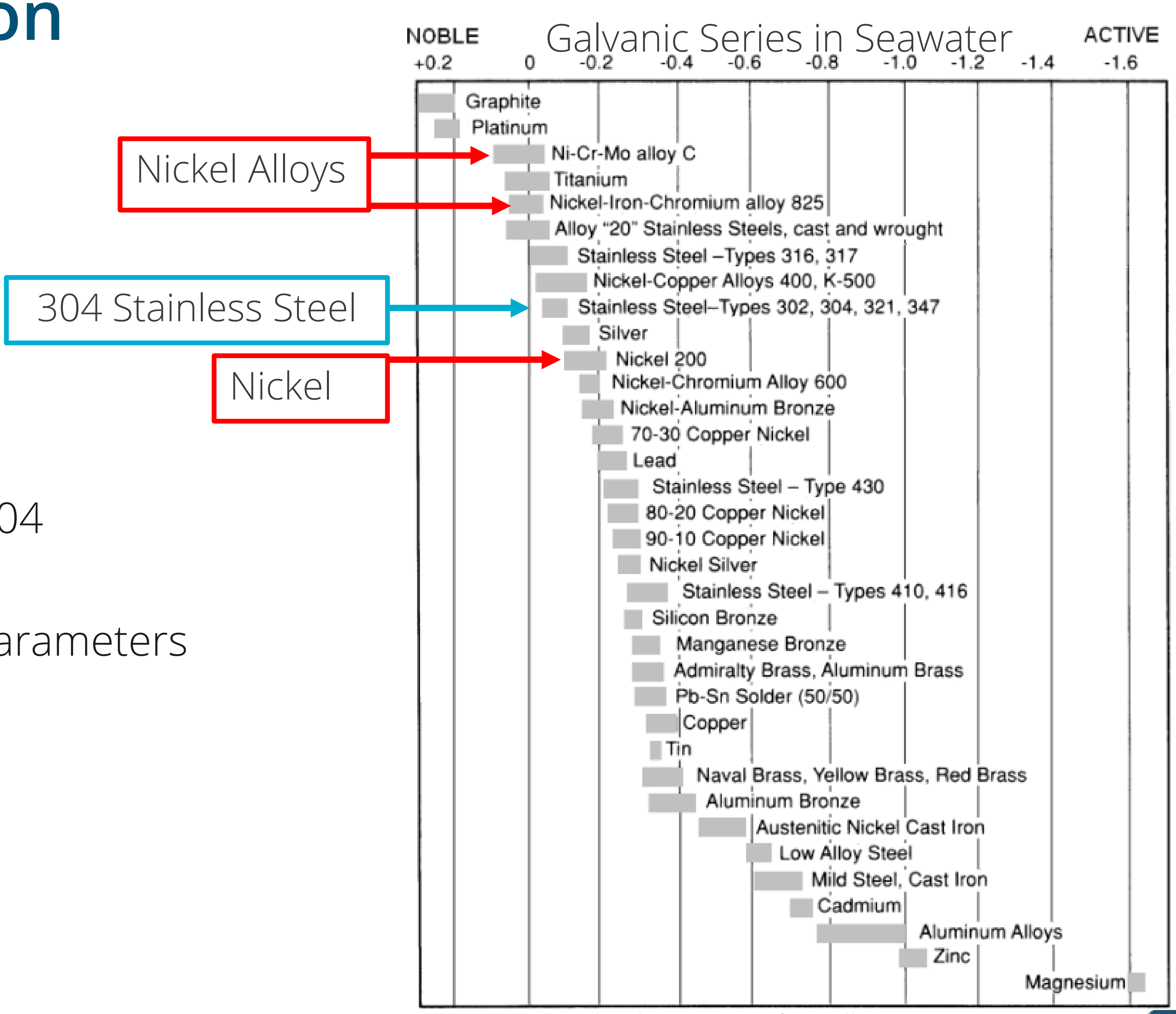
Try to minimize galvanic corrosion through material selection

Started with Nickel, Nickel Alloys:

- Historically good galvanic pair with 304
- Good corrosion behavior
- Existing research: optimized spray parameters

Coating Types:

Inconel 625-He
Inconel 625-N
Super C-N
Nickel-N

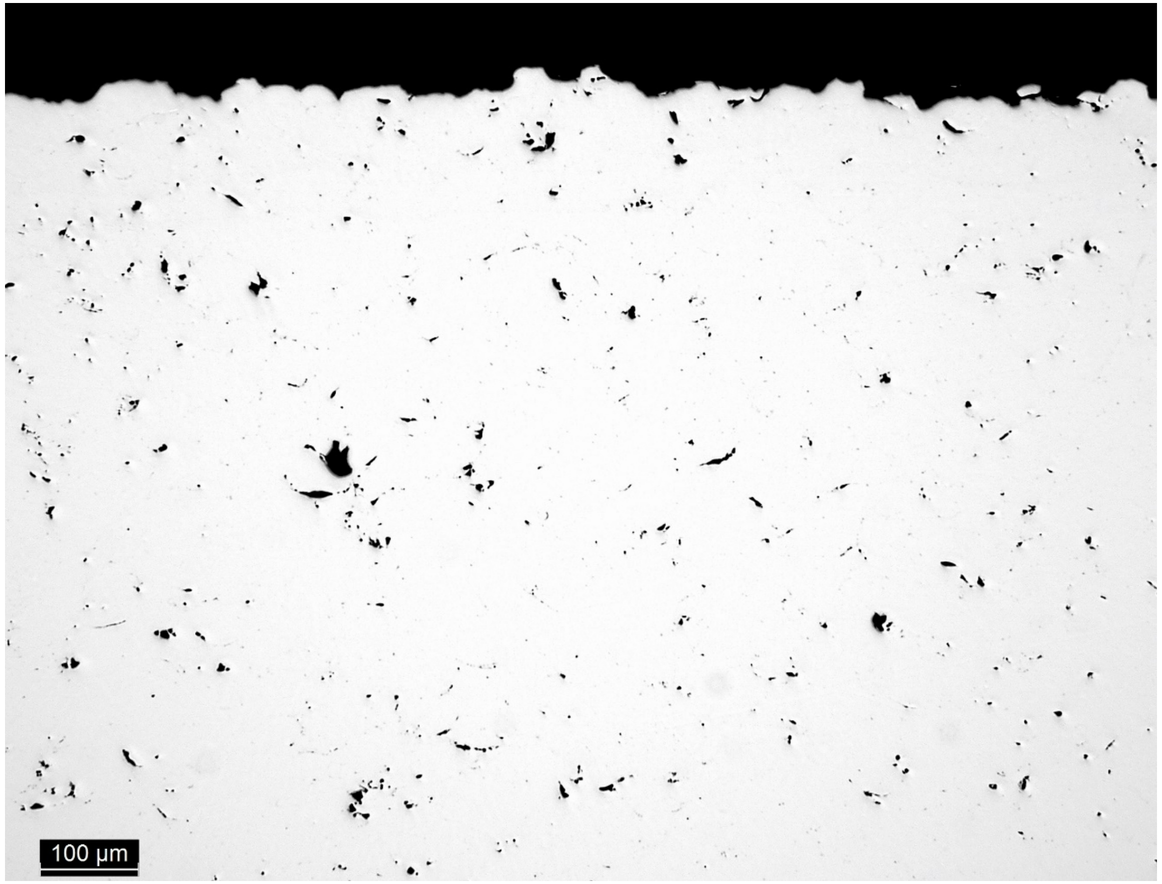


Corrosion Technology- Types of Metallic Corrosion (2013)

Porosity and Roughness

arithmetic mean height (S_a)

Sample	Porosity (%)	S_a (μm)
SC-N	5.51 ± 0.44	16.7 ± 0.5
Inc-He	1.21 ± 0.20	15.7 ± 0.5
Inc-N	5.79 ± 0.18	17.2 ± 0.6
Ni-N	3.78 ± 0.59	18.5 ± 0.6

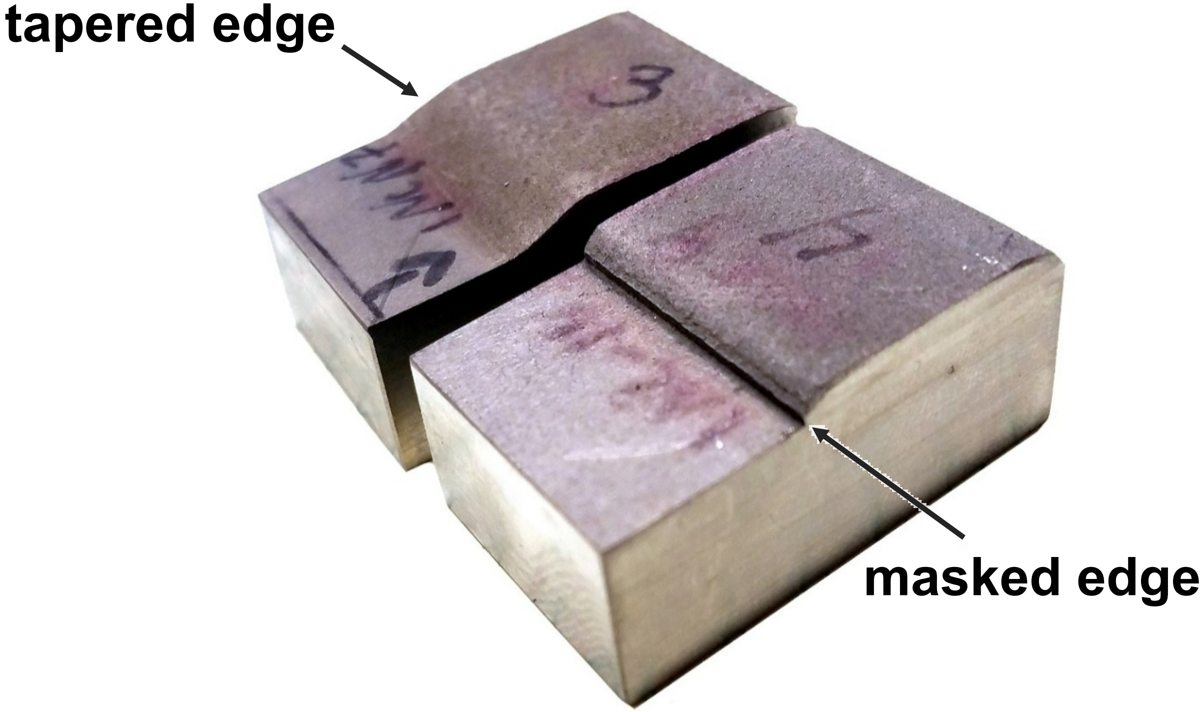


Courtesy of Ken Ross (PNNL)

Cold Spray Samples

Edge types:
- Tapered
- Masked

Edge type will affect:
- Coating coverage at juncture
- Residual stresses at patch edge



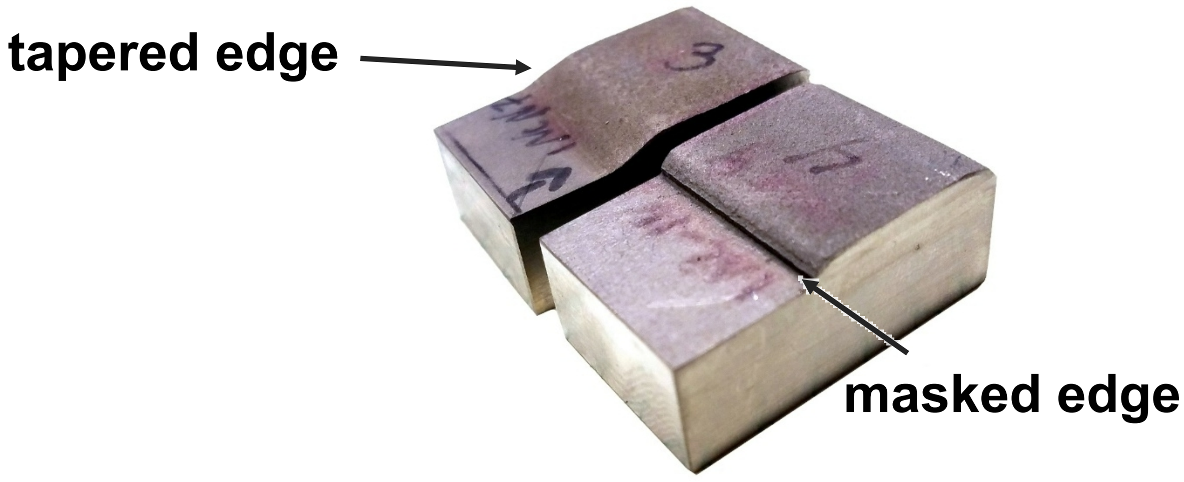
Ross, Kenneth A., et al. *Assessment of Cold Spray Technology for Nuclear Power Applications*. No. PNNL-30299. Pacific Northwest National Lab.(PNNL), Richland, WA (United States), 2021.

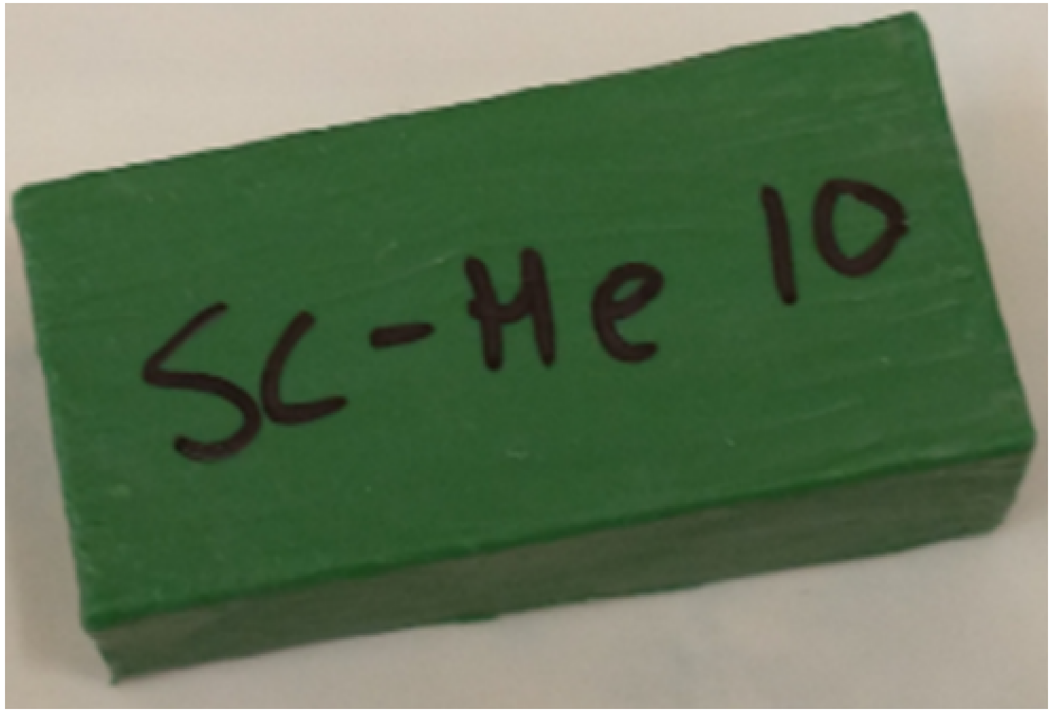
Accelerated Ferric Chloride Testing

Rapid Assessment of Influencing Factors
- Accelerated pitting test
- Evaluate potential galvanic couple influences
- Evaluate interface-type impact

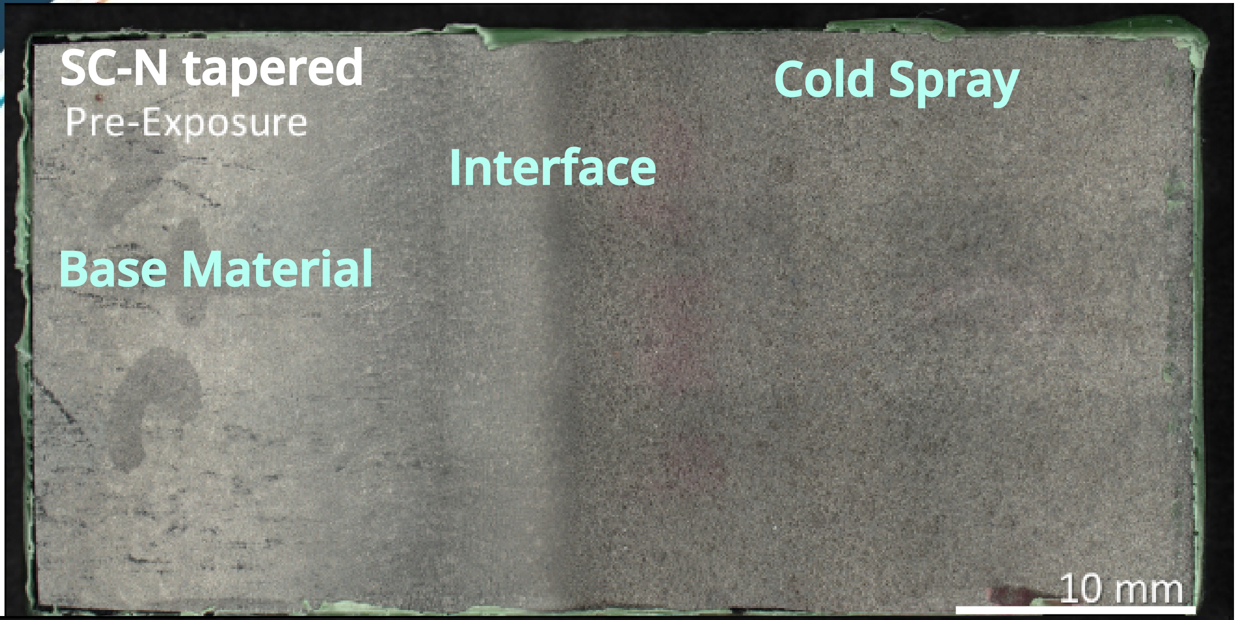
Sample sides/bottom coated

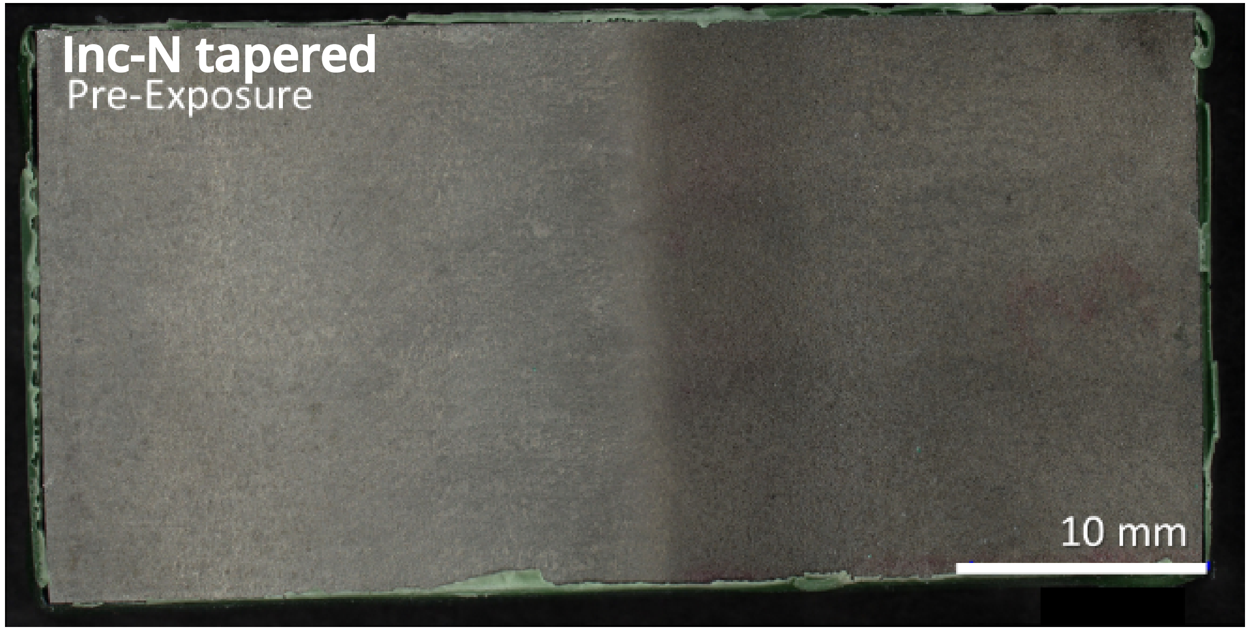
ASTM G48 Method A
Full immersion
6% by mass ferric chloride solution
72 hours at 22 ºC

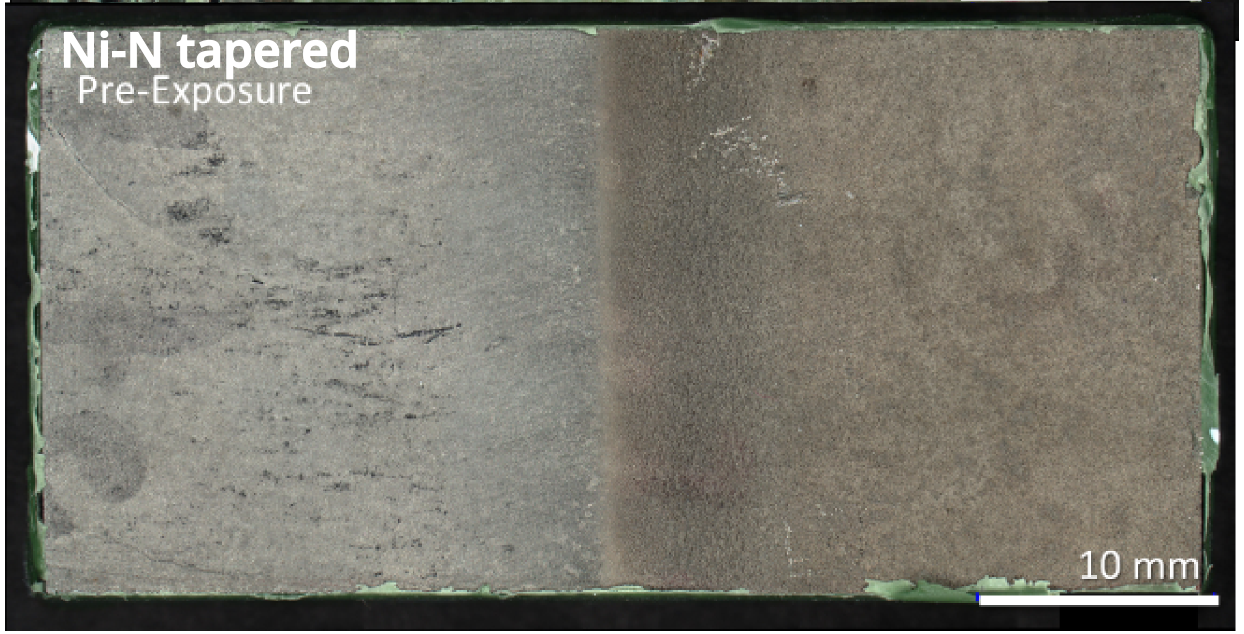




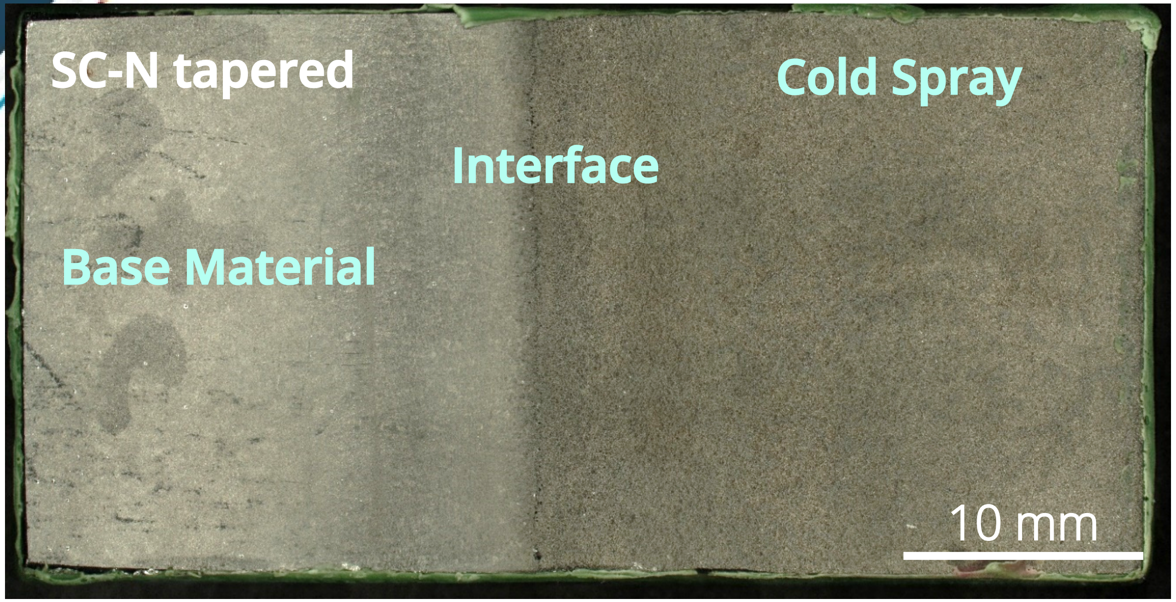
Material Selection: Pre-Exposure

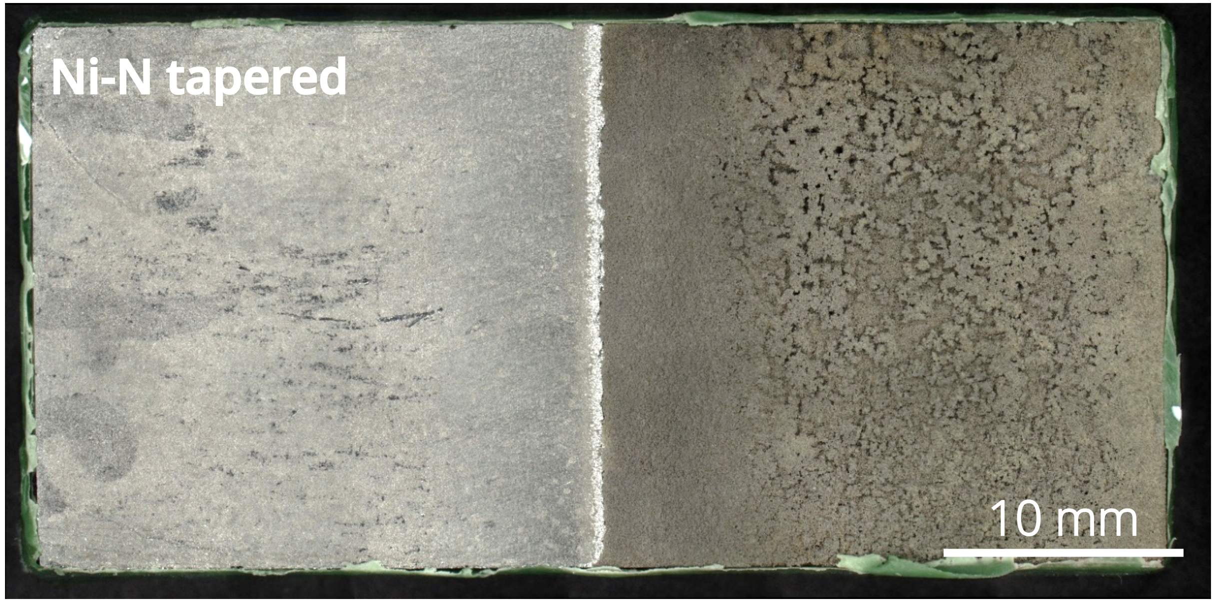






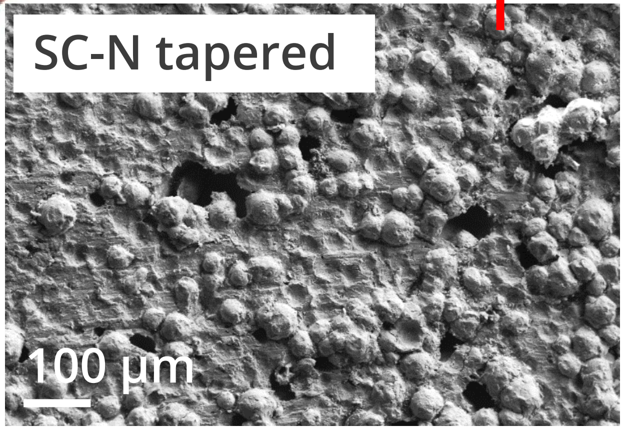
Material Selection: Post-Exposure

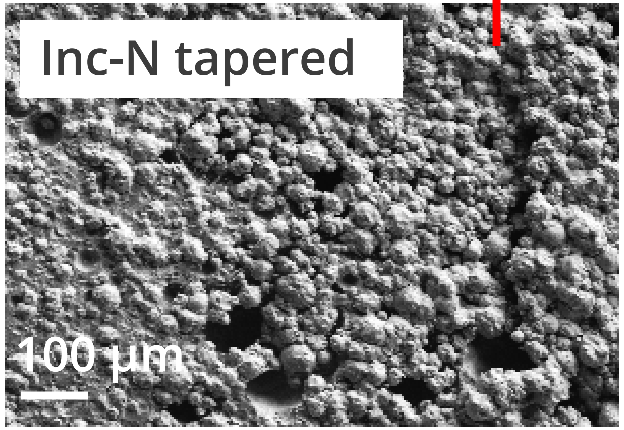


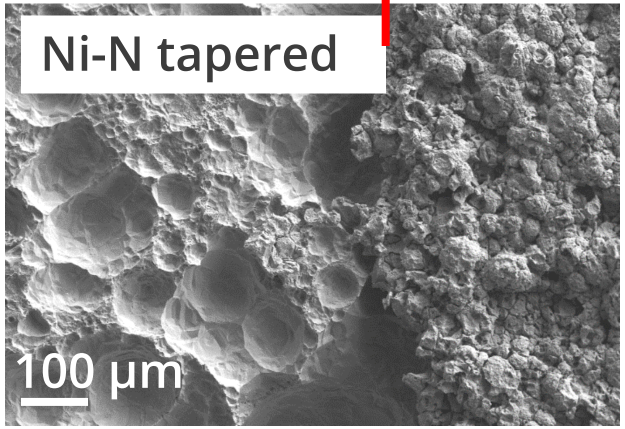


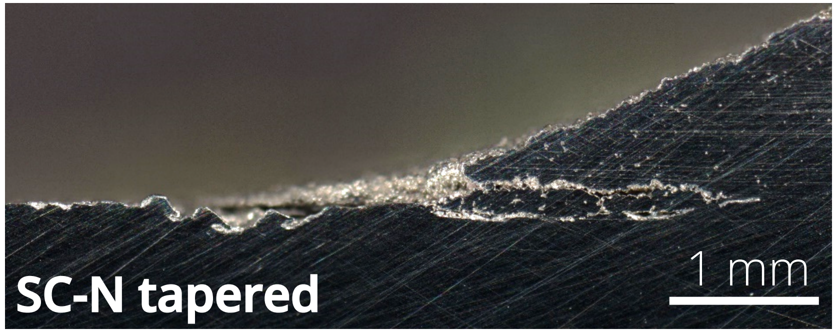
- Ni shows lots of damage in the cold spray & at the interface
- Inc-N & SC-N damage is harder to see

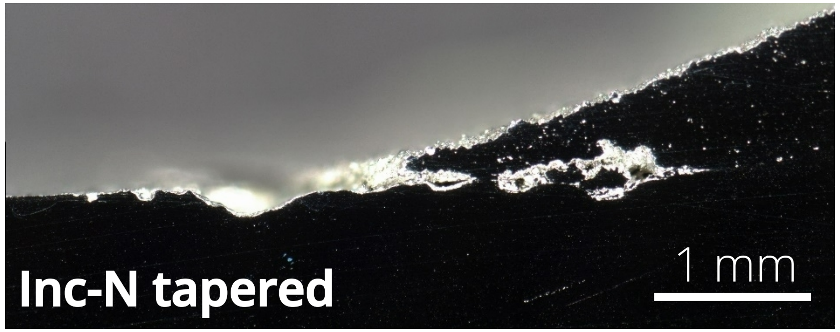
Material Selection

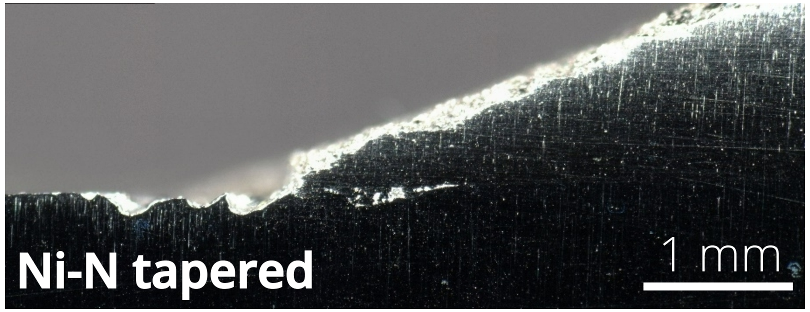












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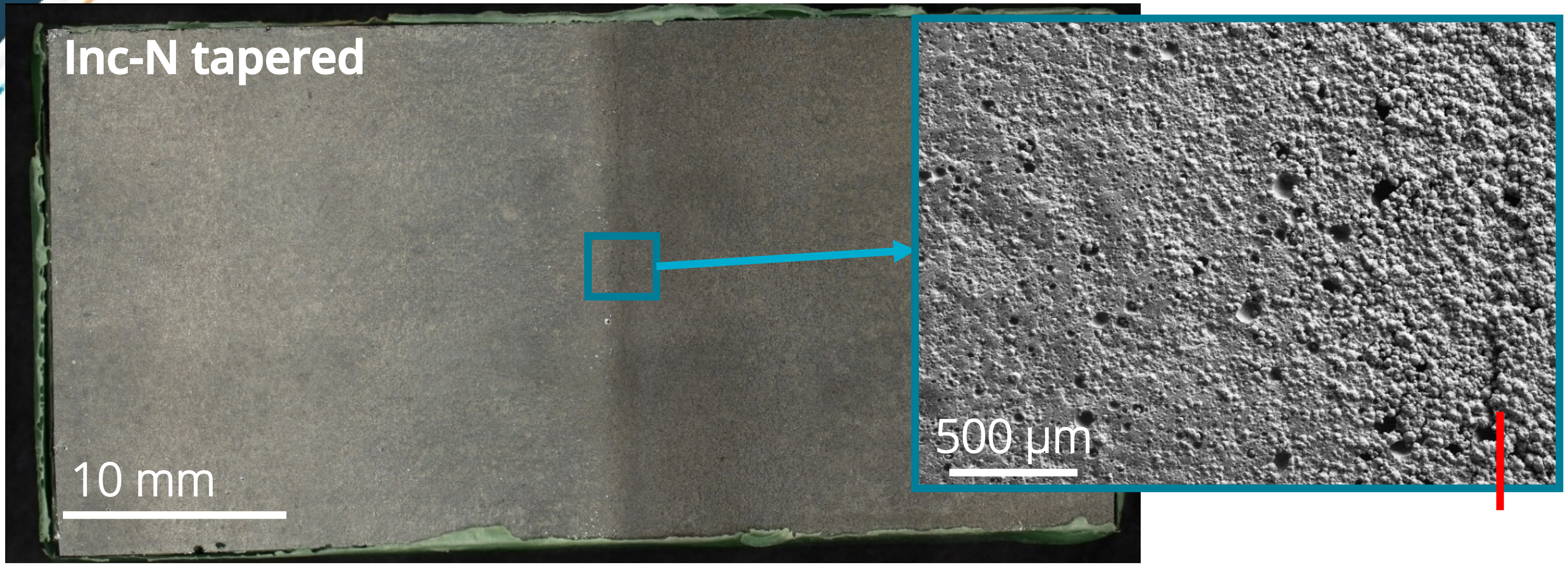
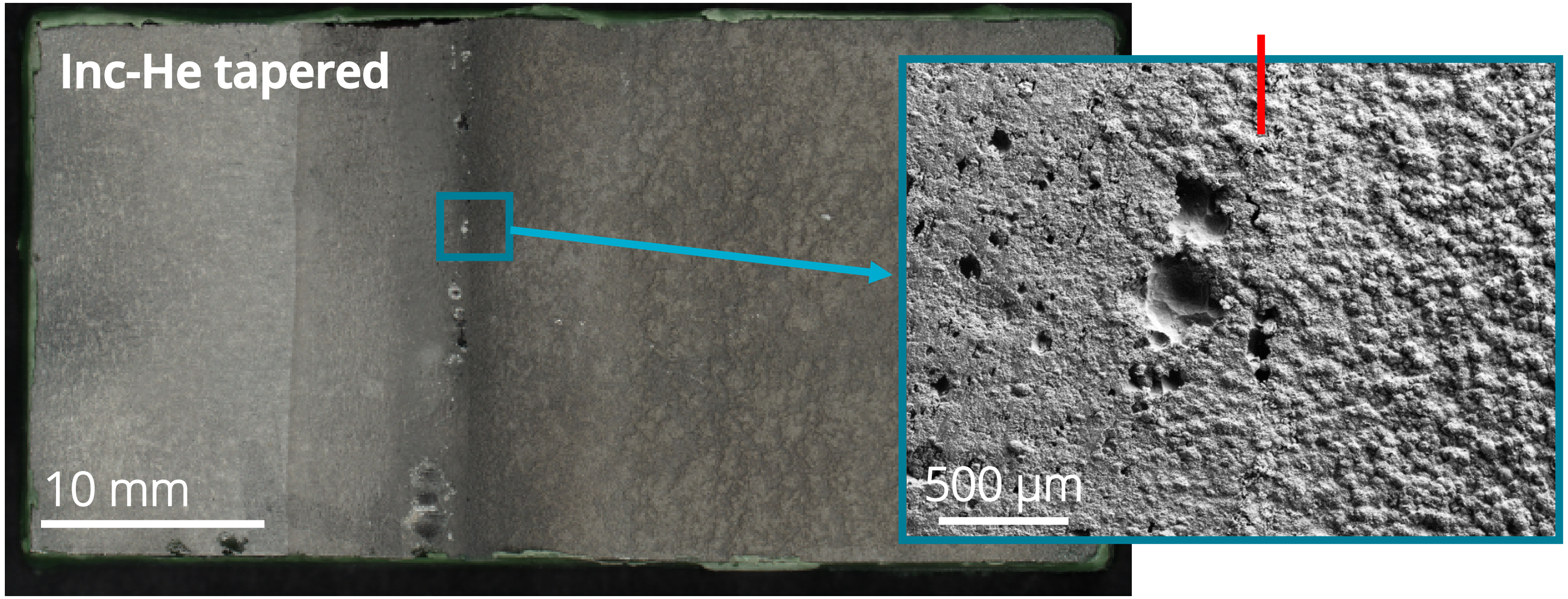
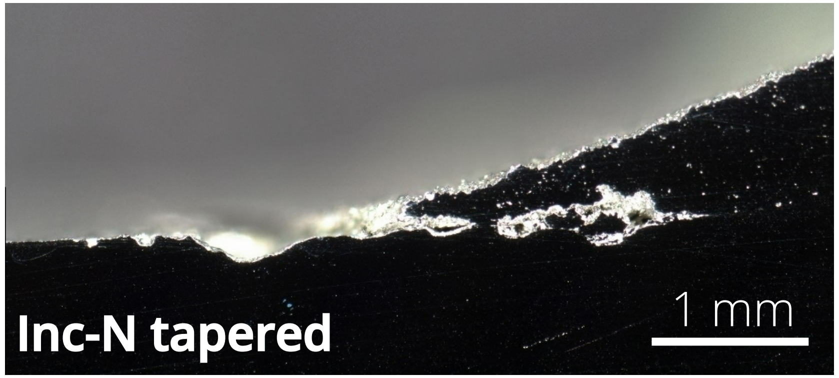
Porosity associated with attack in cold spray layer

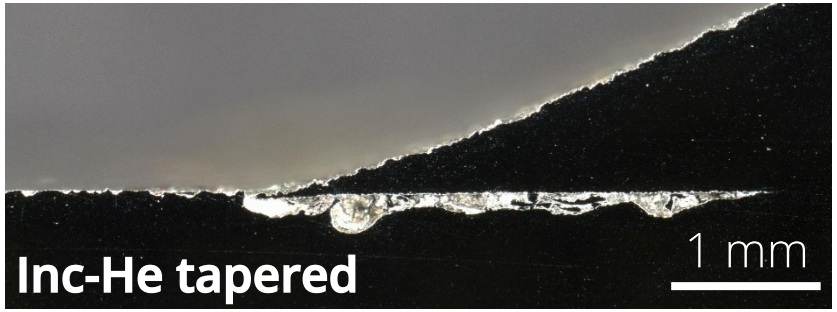
Gas Selection



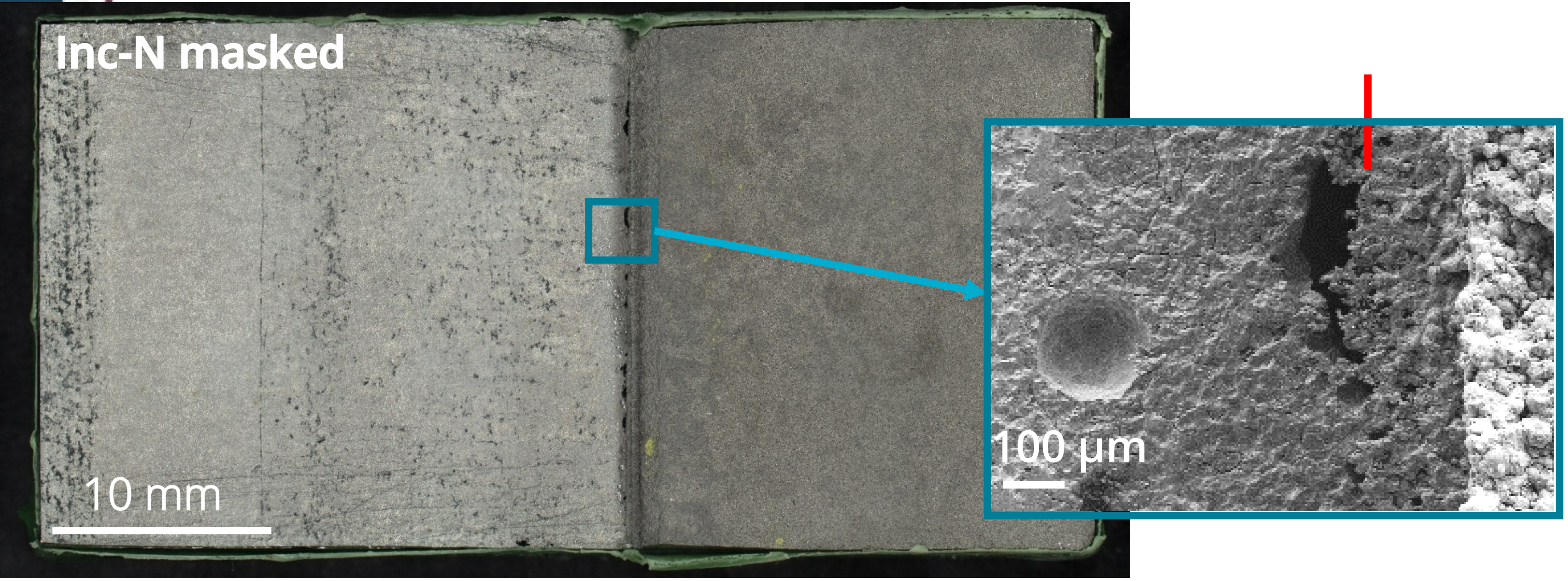


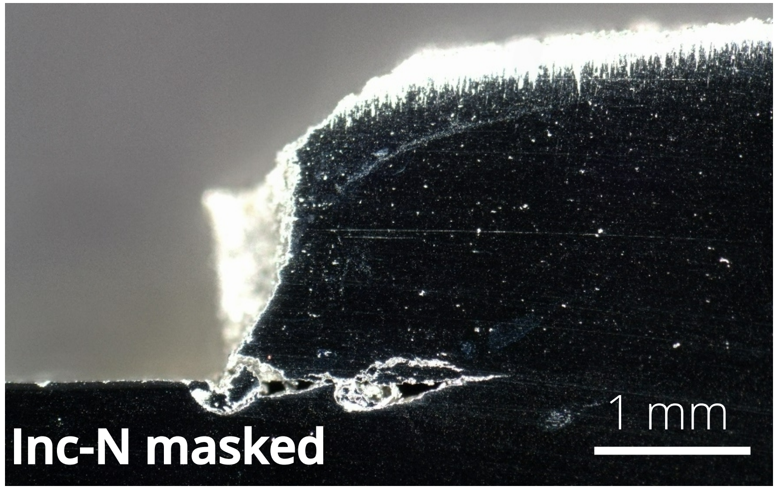


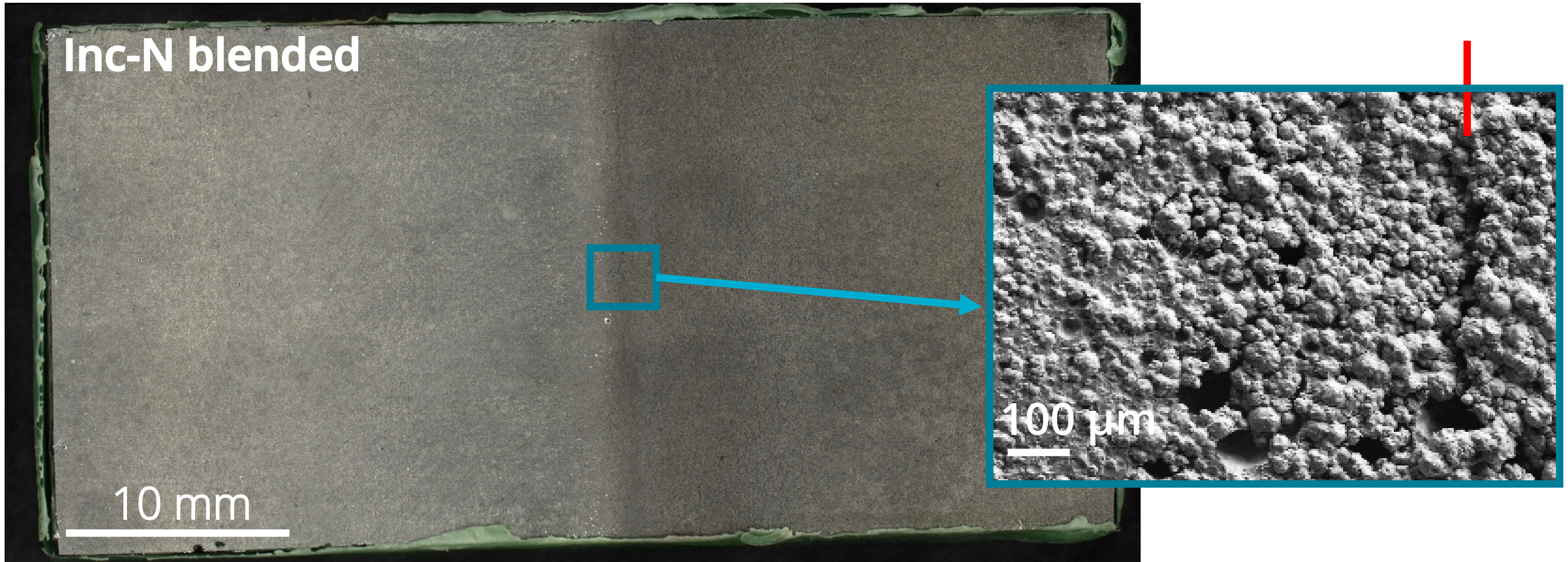
Sample	Porosity (%)
Inc-N	5.79 ± 0.18
Inc-He	1.21 ± 0.20

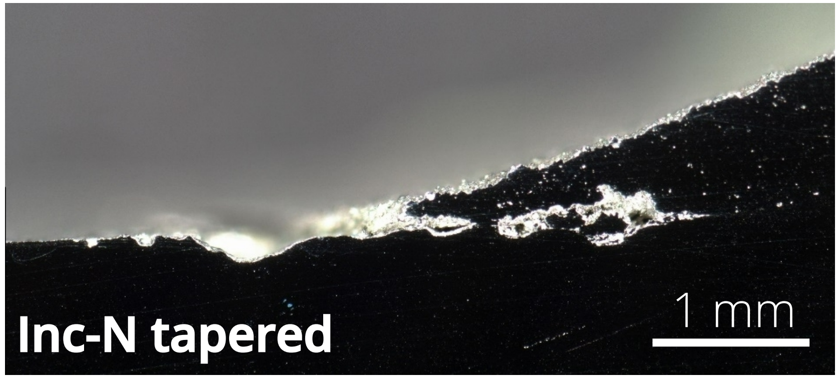


Masked vs Blended: Inc-N



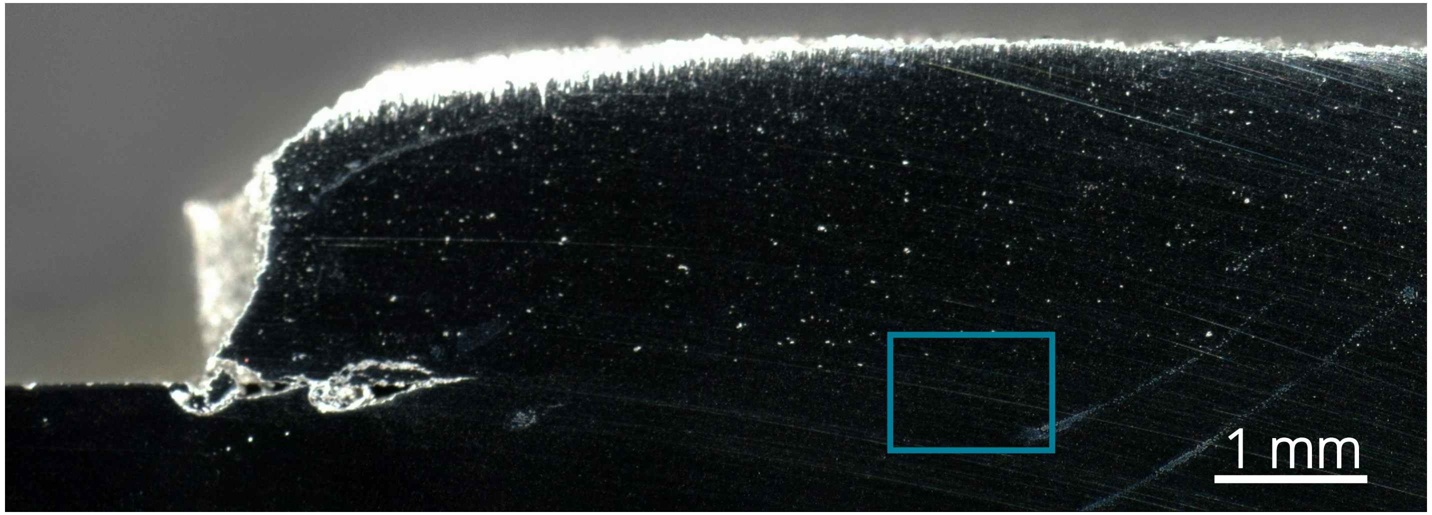






Deformation & Corrosion

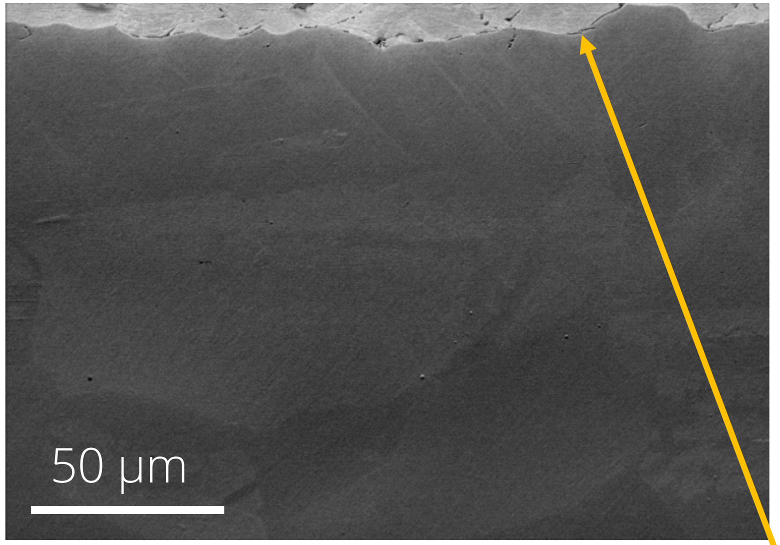
Could a deformation layer be detrimentally advancing corrosion between the cold spray and substrate layers?



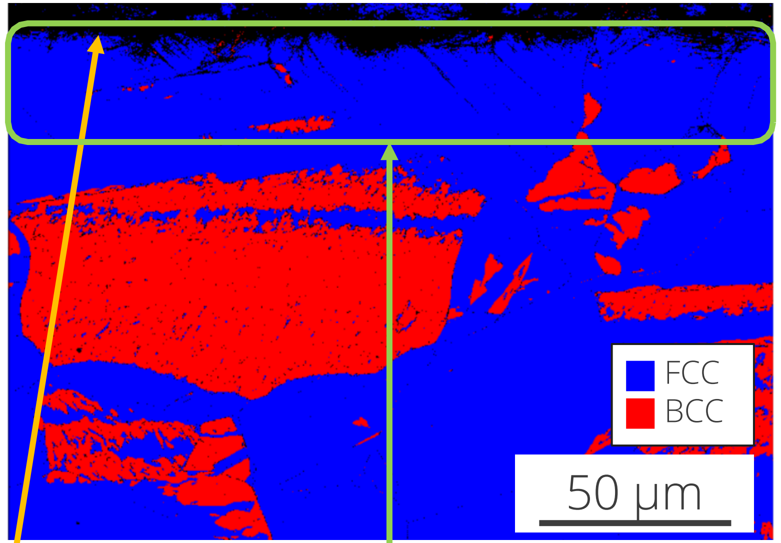
Deformation in stainless steel can reduce corrosion resistance.
In 304 stainless steel, deformation can cause a phase change.
Austenite (FCC) →Martensite (BCC)

Inconel-N Masked: Far from Patch Edge

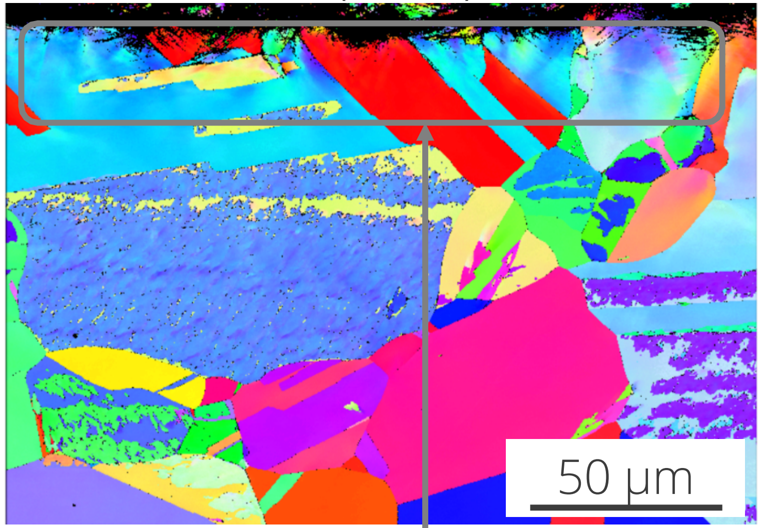
SEM



Phase Map



Electron Backscatter Diffraction (EBSD)

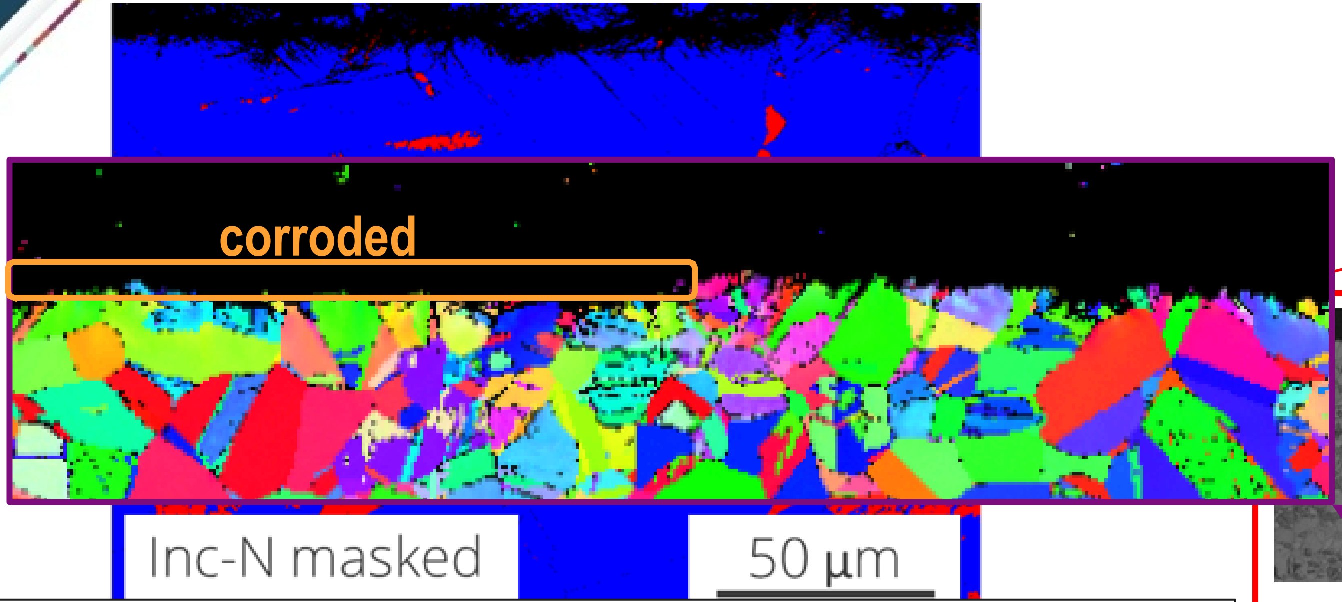


Black band at interface isn't missing material;
Too deformed to get diffraction patterns;

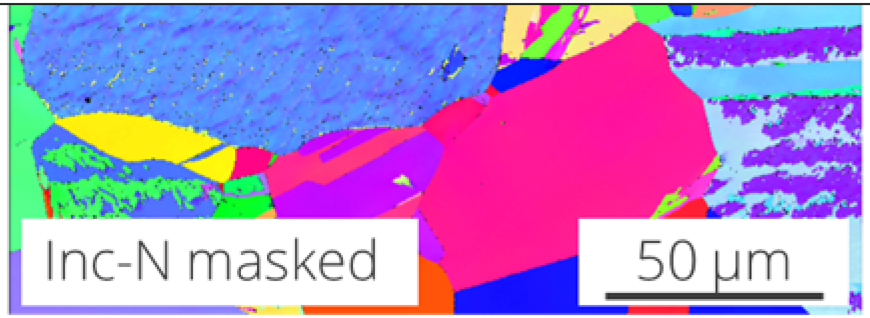
No concentration of BCC at substrate/cold spray interface;
BCC phase consistent with bulk;

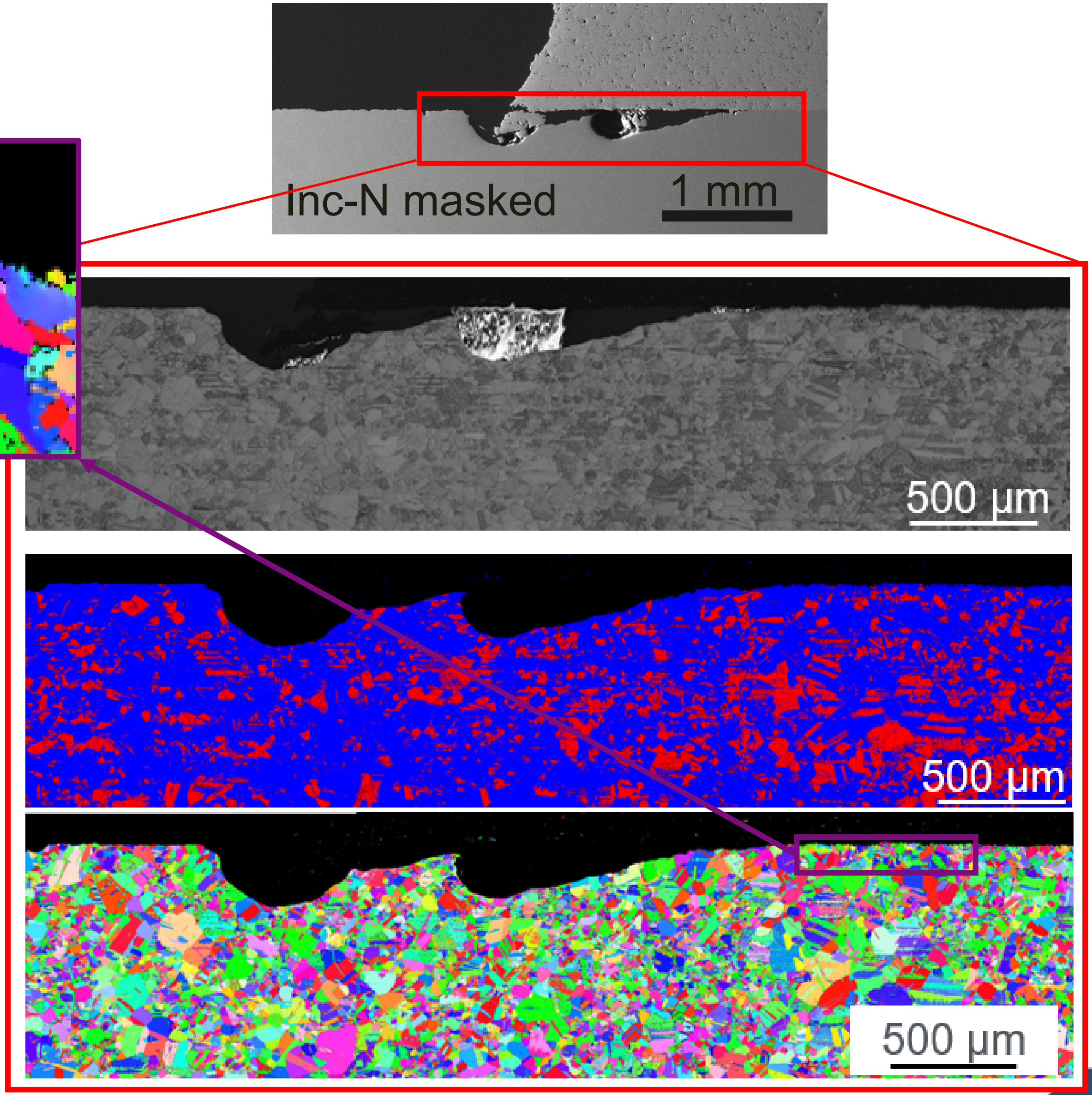
Water-colory region indicates deformation;
~25 µm deep

Corrosion in Deformation Layer



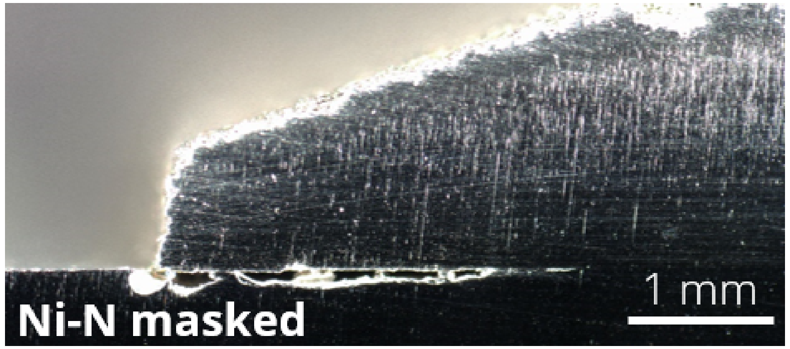
Corrosion seems to travel more rapidly through deformed layer. Eventually corrosion spreads beyond it.

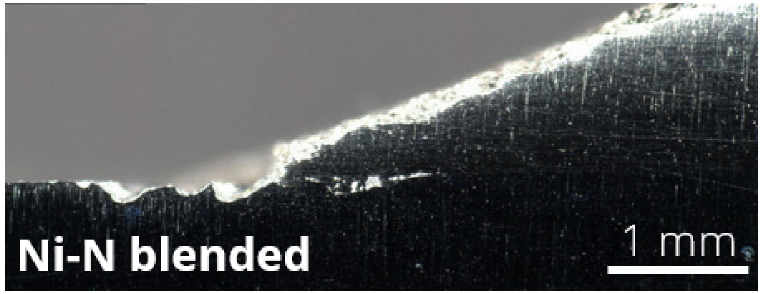


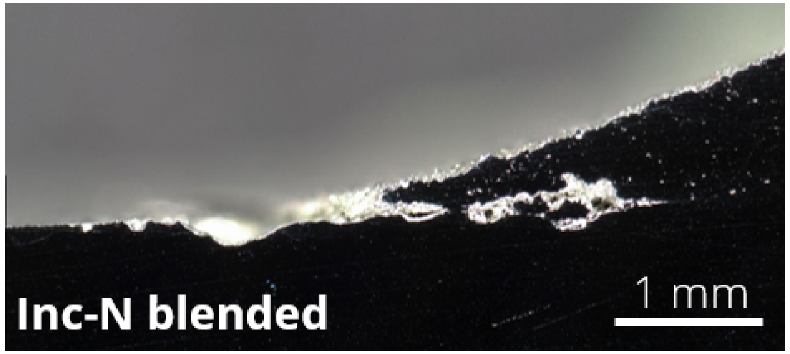


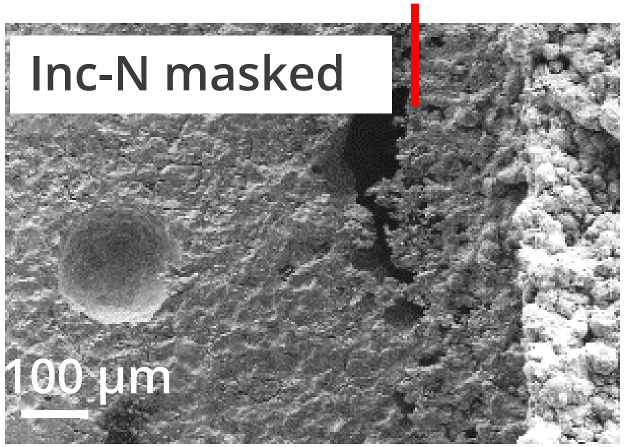
Ferric Chloride Testing Summary

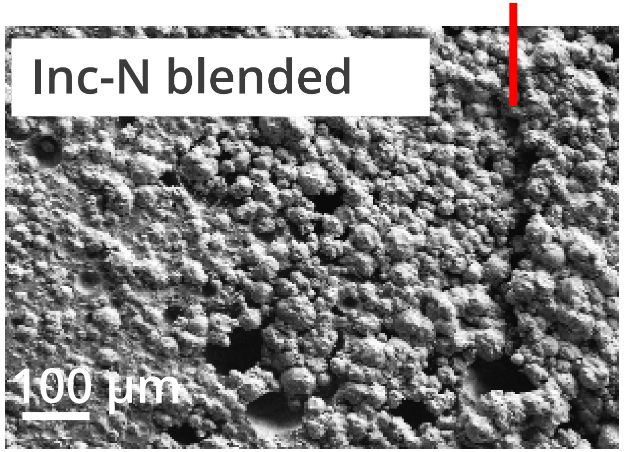
- Damage morphology differs with interface type
- Surface roughness obscures extent of attack
- Top-down damage ≠ corrosion under the cold spray
- Porosity influences location of corrosion damage
- Deformation layer thinner than damage











Atmospheric Corrosion

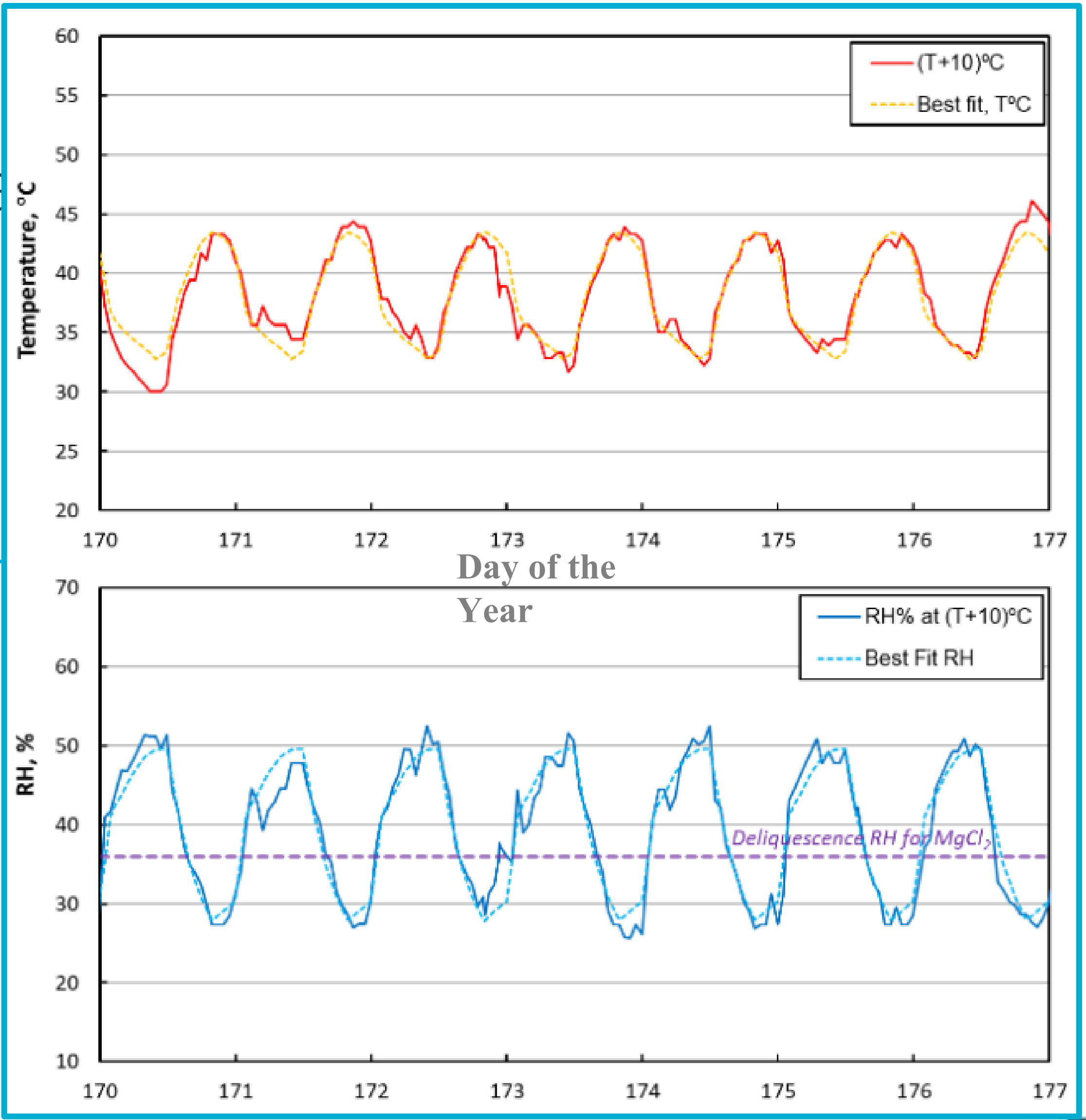
Printed 300 μg/cm² of Artificial Sea

Relative Humidity:

- 40% RH; 35 °C
- 75% RH; 35 °C
- cyclic

Above $MgCl_2$ deliquescence

Above NaCl deliquescence in ASW



developed from Arkansas Nuclear 1 ISFSI (SNL 2021 M2)

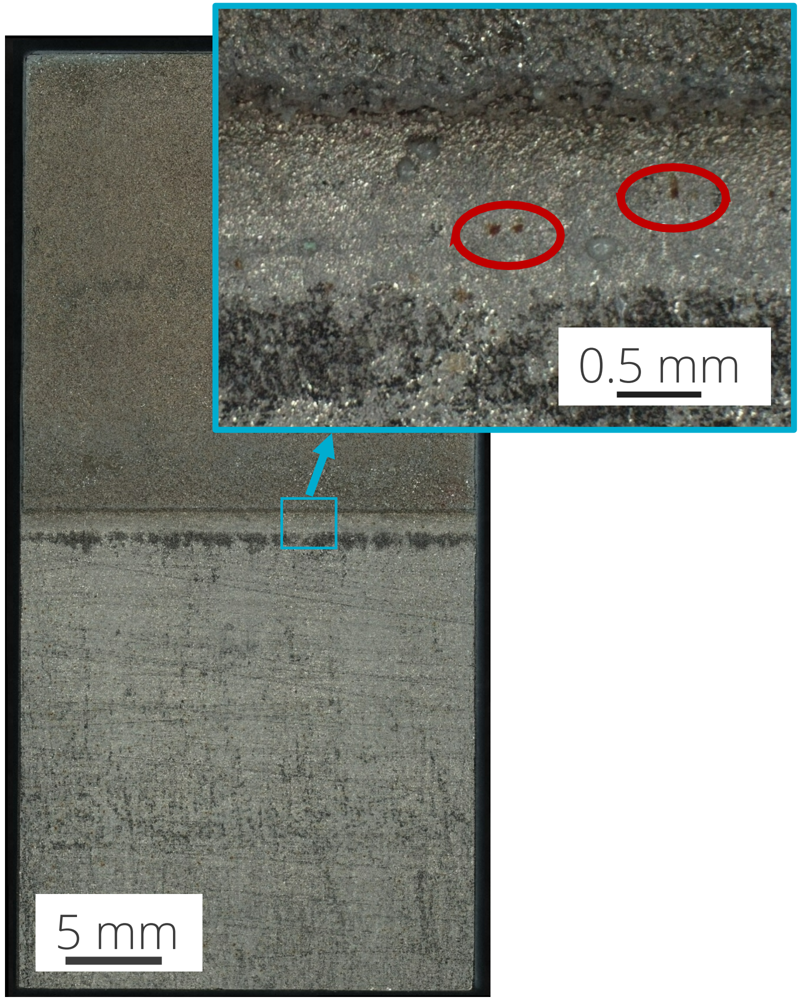
Ni-N Masked: Pre-Exposure

40% RH

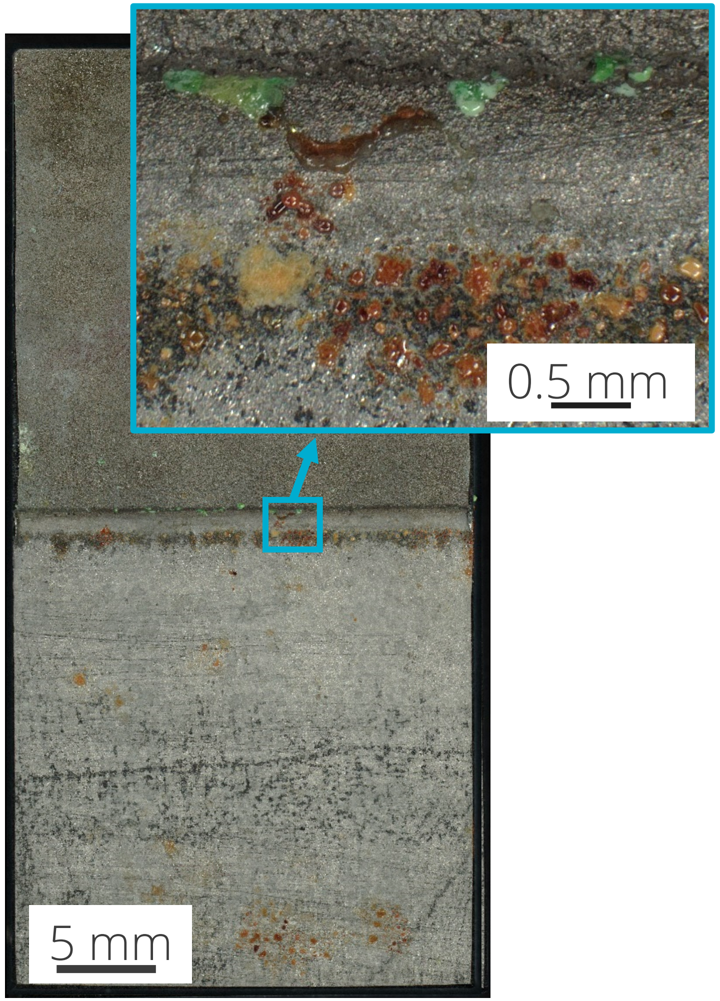
75% RH

cyclic

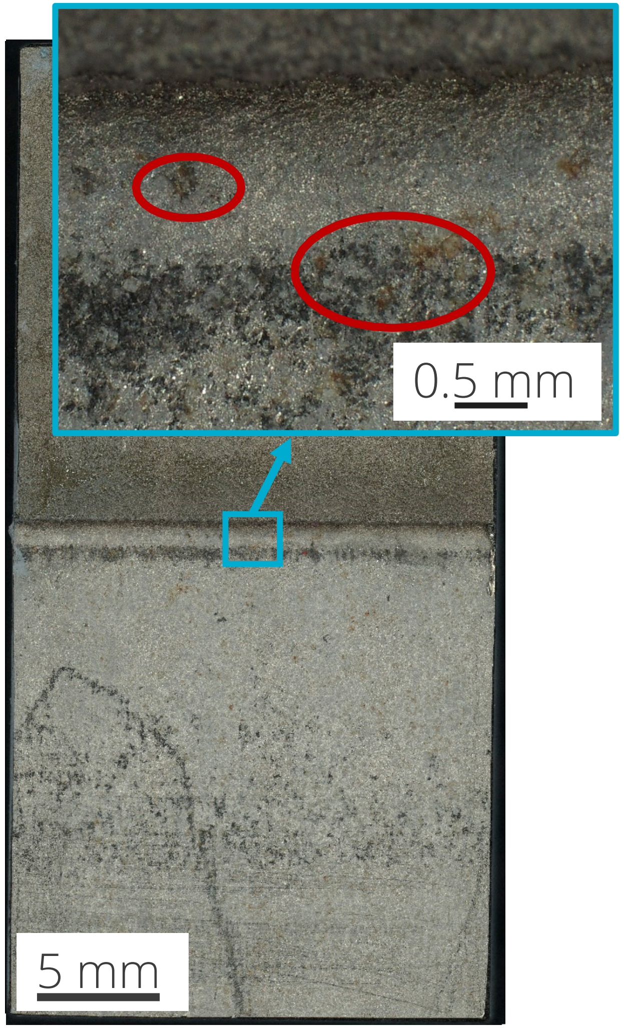
Ni-N Masked: 1 Month Exposure



40% RH

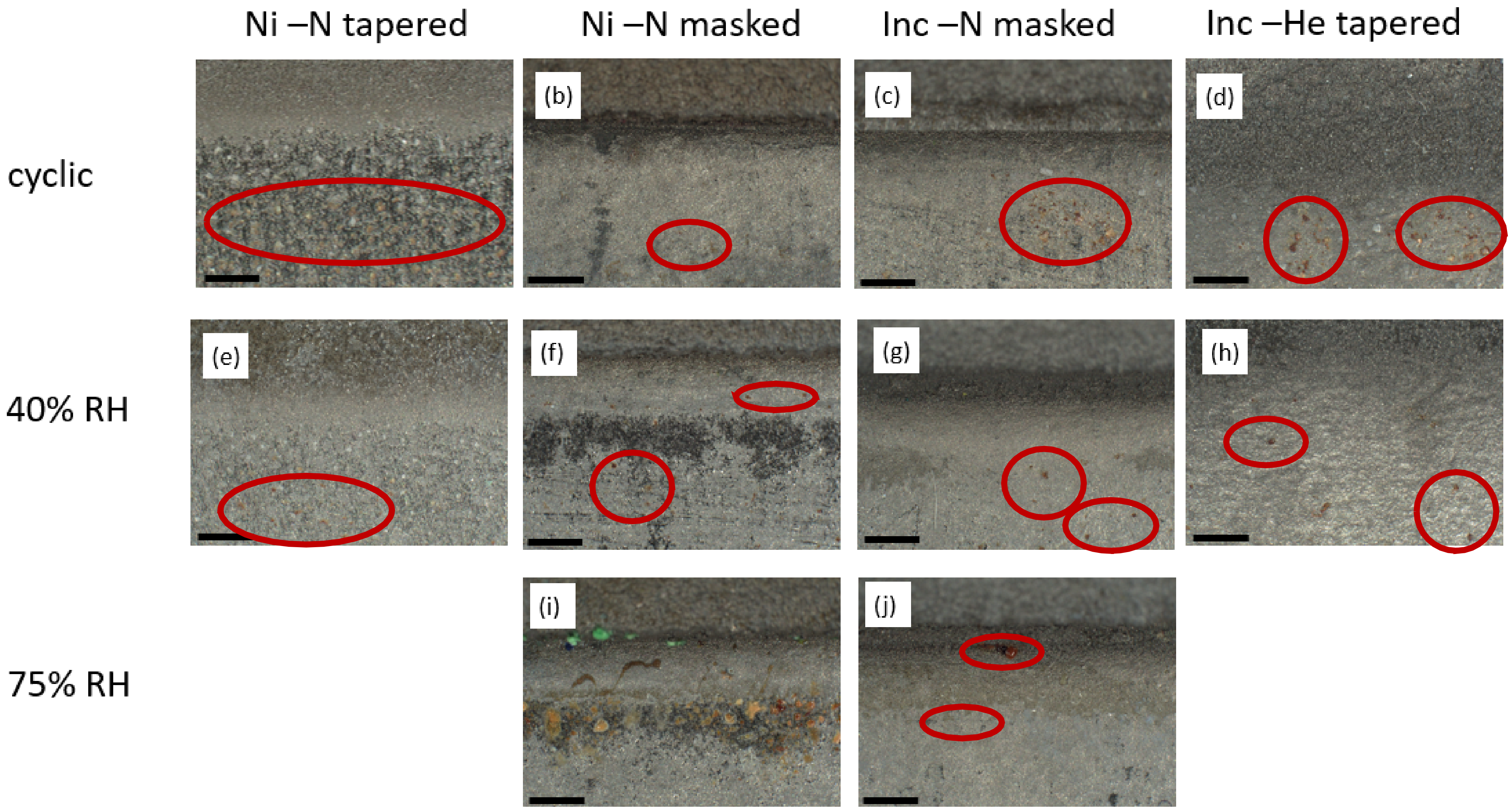


75% RH



cyclic

Atmospheric Corrosion: 8 Week Exposure



Corrosion location similar to that observed in Ferric Chloride Testing

Summary

Materials Selection:

- galvanic influences

Processing:

- gas selection
- interface selection

Deformation:

- microstructural transformation
- residual stresses

Behavior in Realistic Environments

- **long term**
- **different brine chemistries**
- **different stress-states**

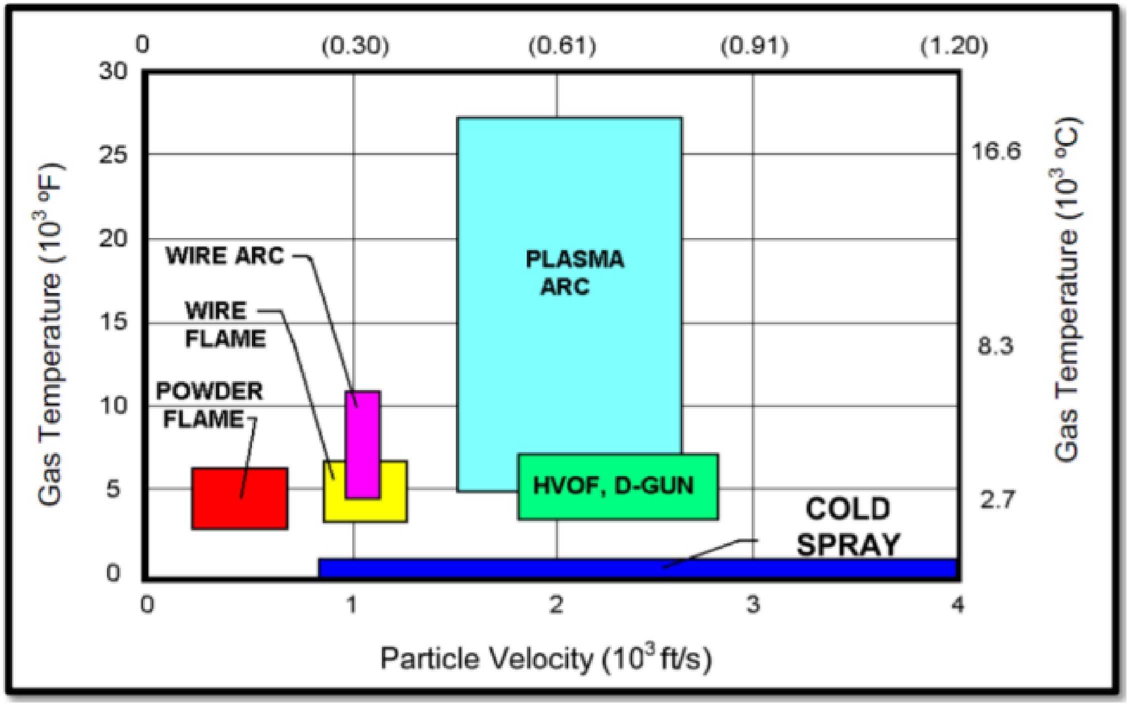
Questions?

Acknowledgements:
Rebecca Schaller, Jason Taylor, Timothy Montoya, Charles Bryan
Sandia National Laboratories
Ken Ross
Pacific Northwest National Laboratory

Cold Spray Applications

Lower temperature helps prevent:

- Temperature-dependent crystalline transformations
- Oxidation
- Vaporization



M. Neshastehriz, 2014, Pennsylvania State University

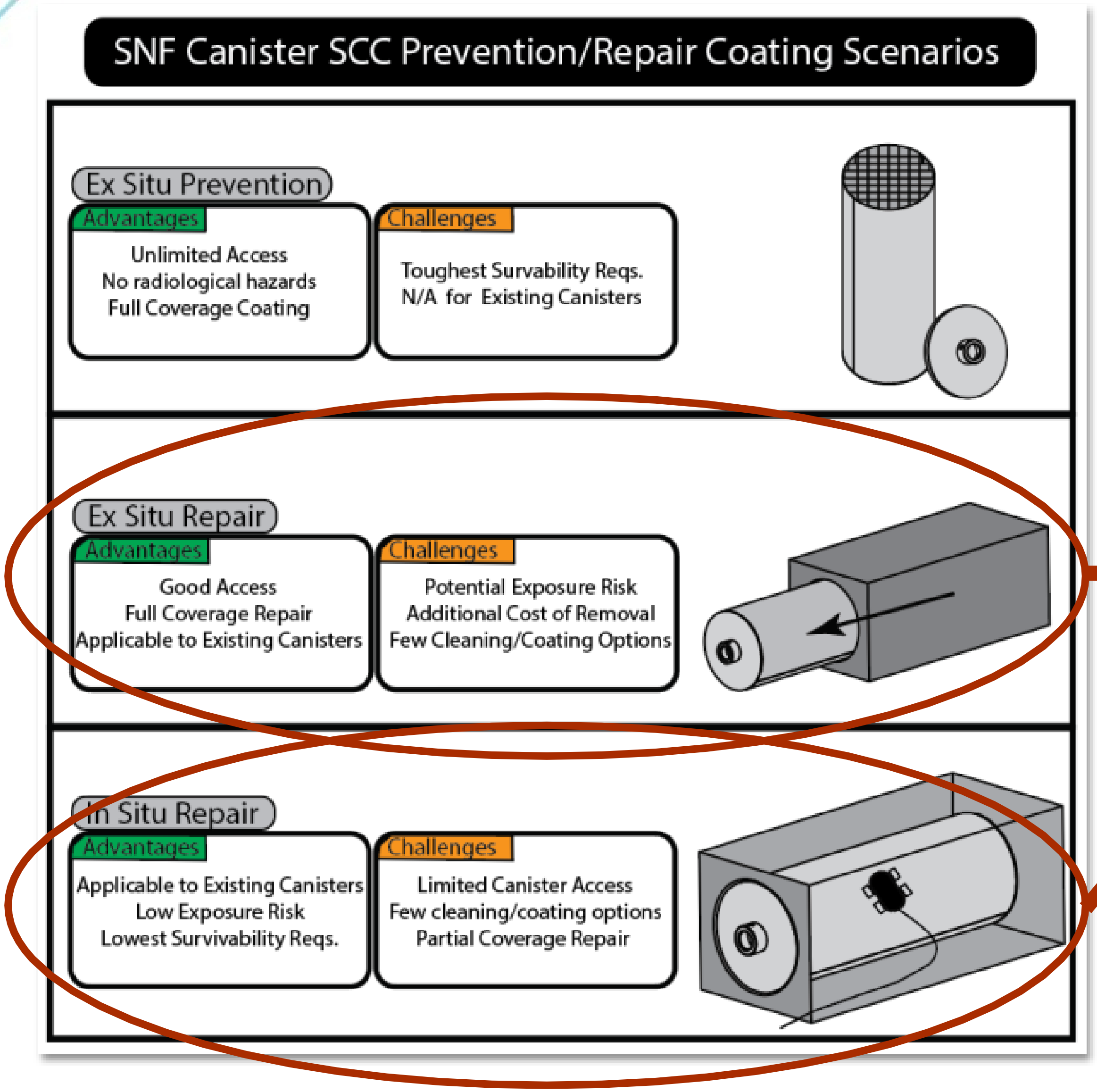
Functions:

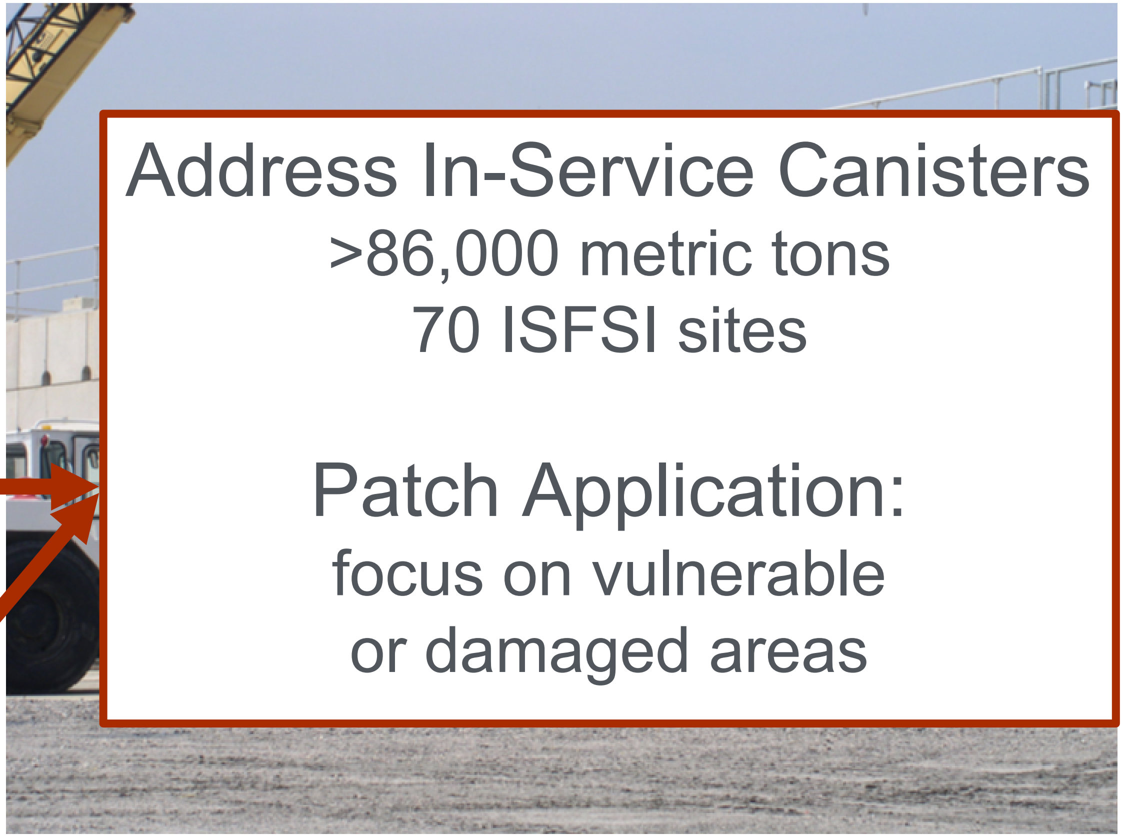
- Additive manufacturing light weighting
- Structural repairs
- Temperature protection coating
- Weld protection coating
- Corrosion prevention/repair coating

Applications:

- Medical devices
- Aircraft
- Automobiles
- Nuclear power

Canister Coating Scenarios



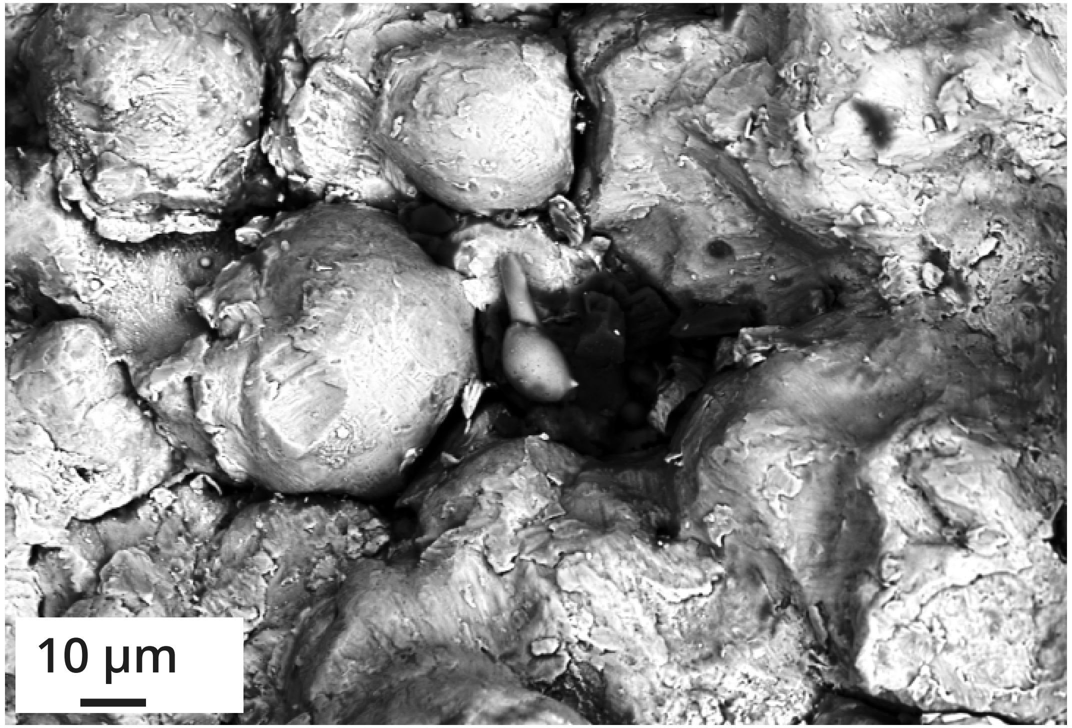


Loading of NUHOMS Storage Cask (TN International, Orano)

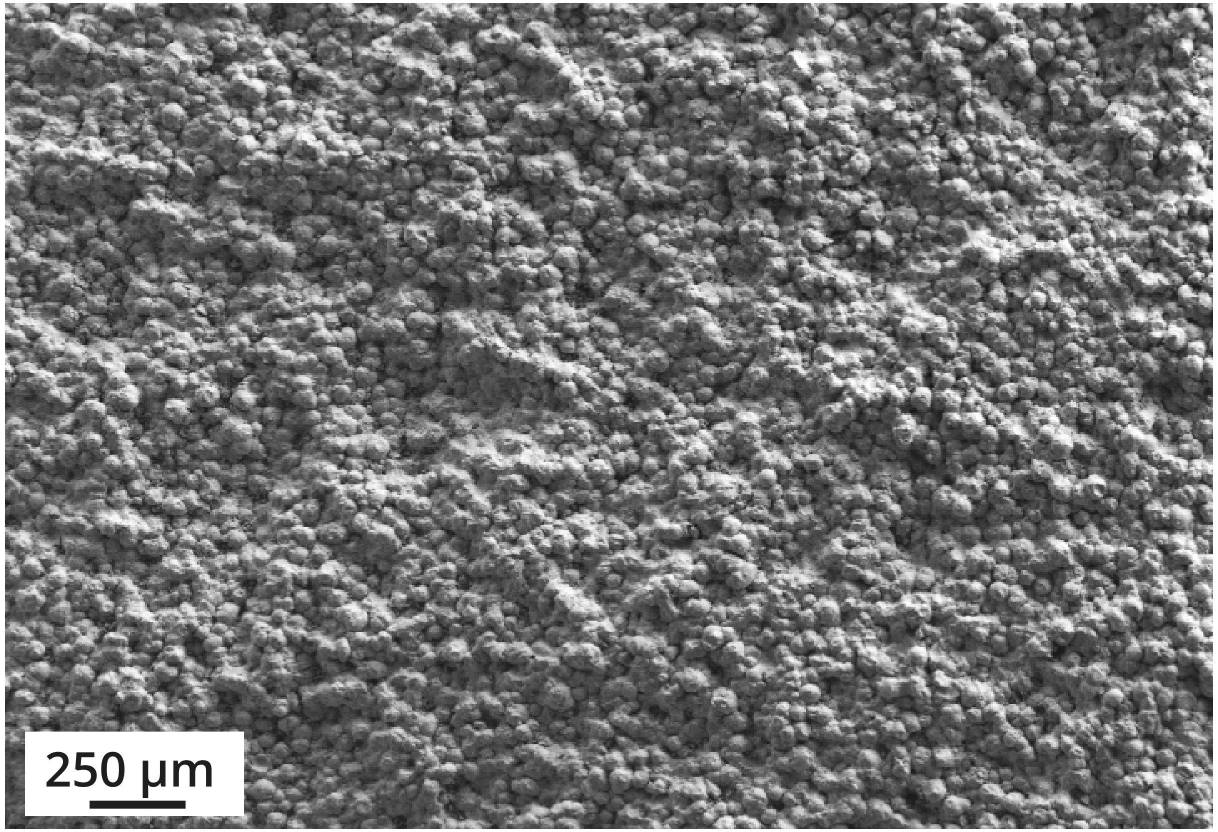
Corrosion Concerns- Metal Particles

Common Issues:

- Porosity
- Surface Roughness



Porosity



Surface Roughness

Cold Spray Coatings

	Inconel 625 (Inc)	Super C (SC)	Commercially Pure Nickel (Ni)
C	0.02%	0.02%	≤0.01%
Co	0.09%	0.2%	-
Cr	21.47%	23.2%	-
Ni	Balance	Balance	>99.9%
Mo	9.06%	17.7%	-
Mn	0.04%	0.7%	-
P	0.0%	0.002%	-
S	0.0%	0.004%	≤0.001
Si	0.07%	0.5%	-
Fe	4.62%	0.6%	≤0.14%
Al	0.04%	-	-
B	0.001%	0.003%	-
Nb	3.65%	-	-
O	0.024%	-	≤0.4%
V	-	0.30%	-
W	-	0.26%	-

Accelerating Gas:
- Nitrogen
- Helium

Affects Porosity
Cost/Sustainability

Coating Types:

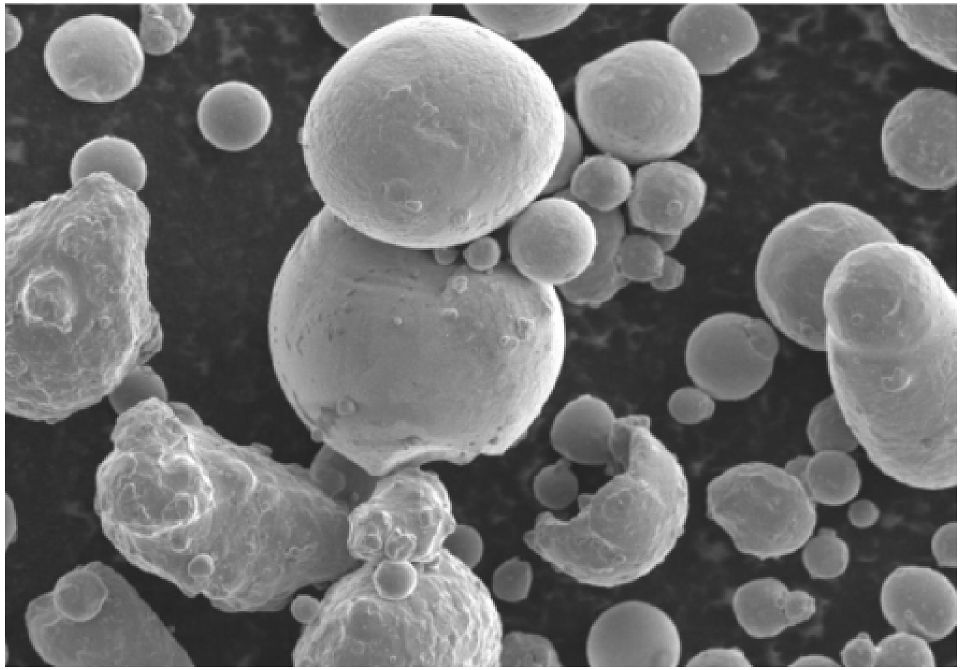
Inc-He
Inc-N
SC-N
Ni-N

Types of Testing

Rapid Assessment of Influencing Factors

Accelerated Testing:
- Electrochemical Scans
- Ferric Chloride Pitting Test

Relevant Conditions:
- Atmospheric Exposures (preliminary)

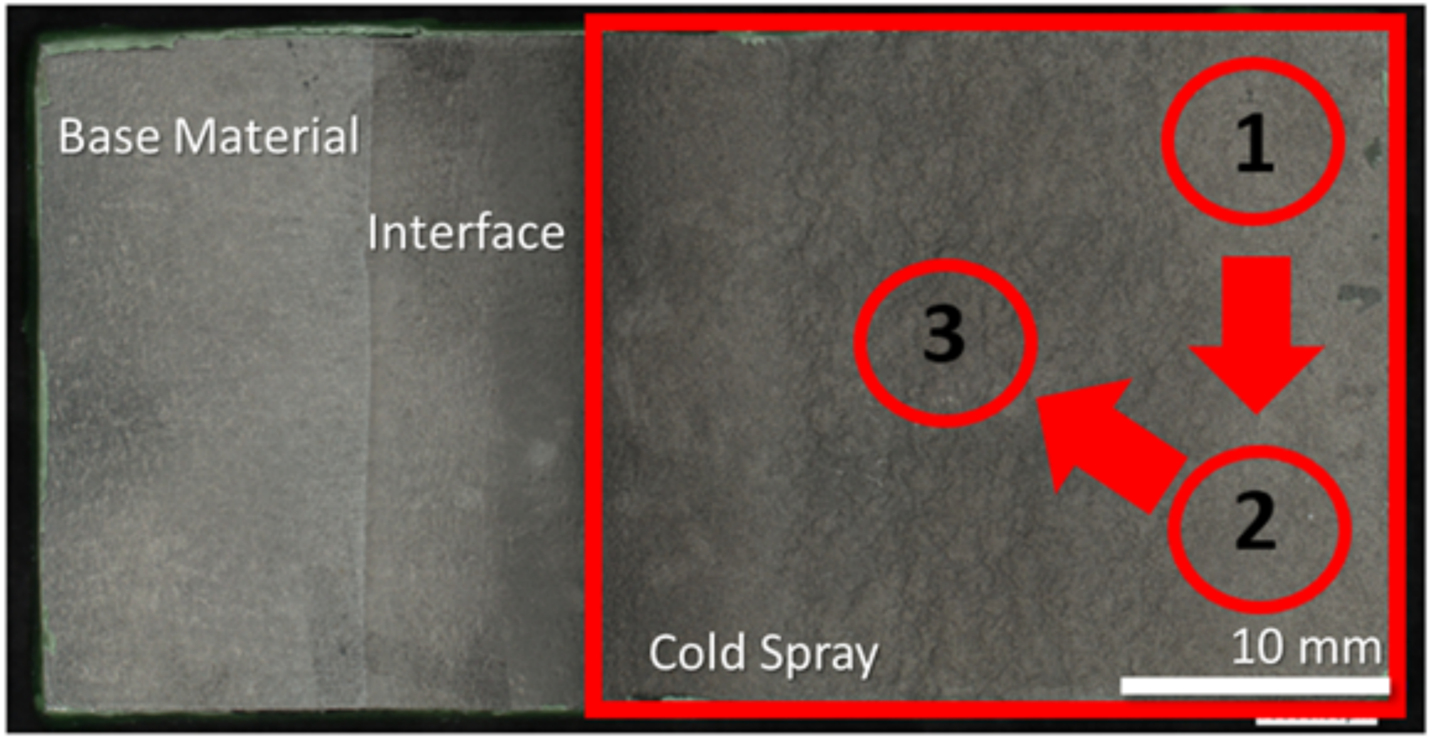


316L stainless steel powder

Electrochemical Scans

Kanopy Tech

Electrolyte: 0.6 M NaCl
Counter Electrode: platinum mesh
Reference Electrode: Ag/AgCl



1 hour Open Circuit Potential (OCP)
Potentiodynamic Scan

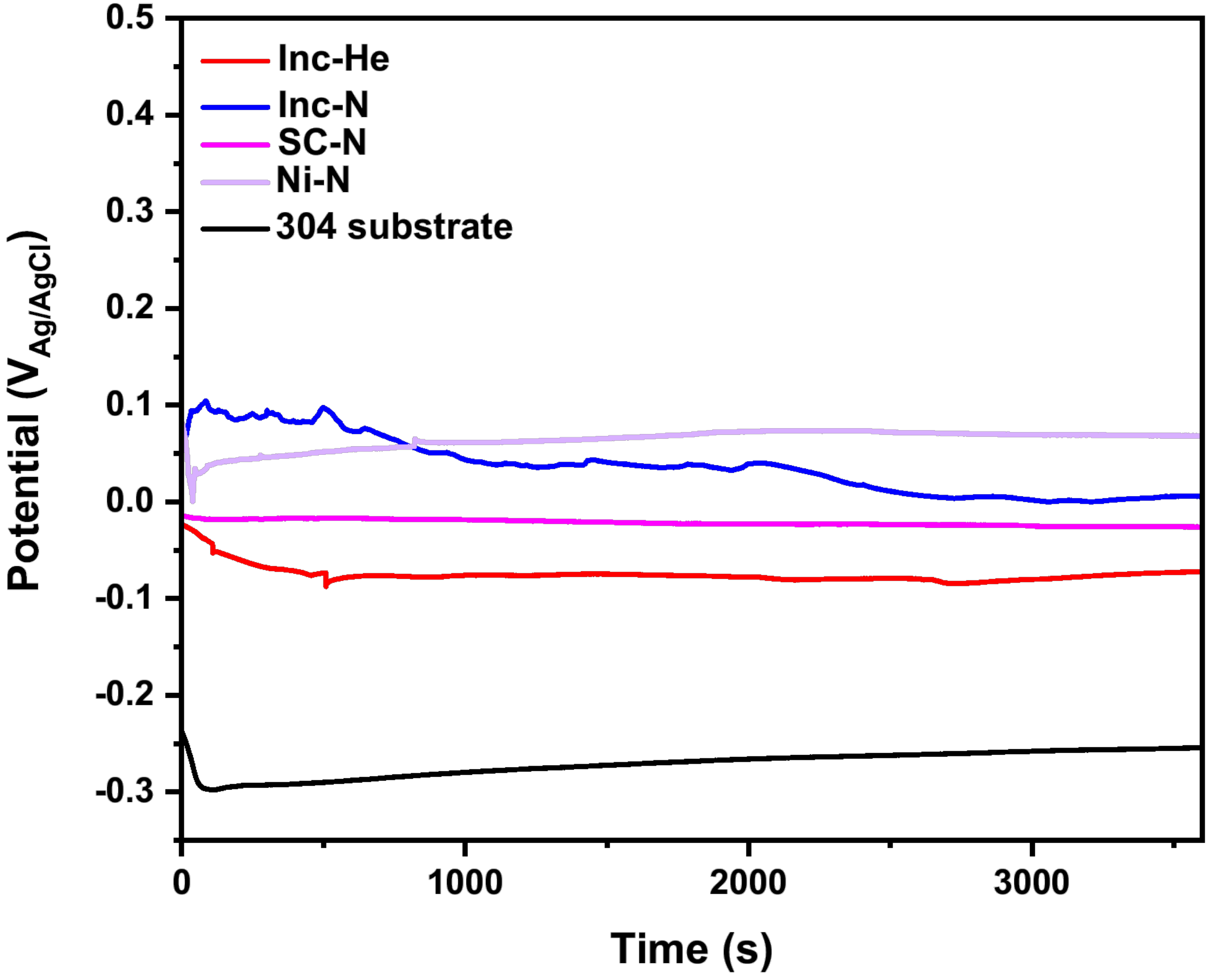
3 spots per sample
Finishes:

- As-Sprayed
- Ground (600, 1200 grit)

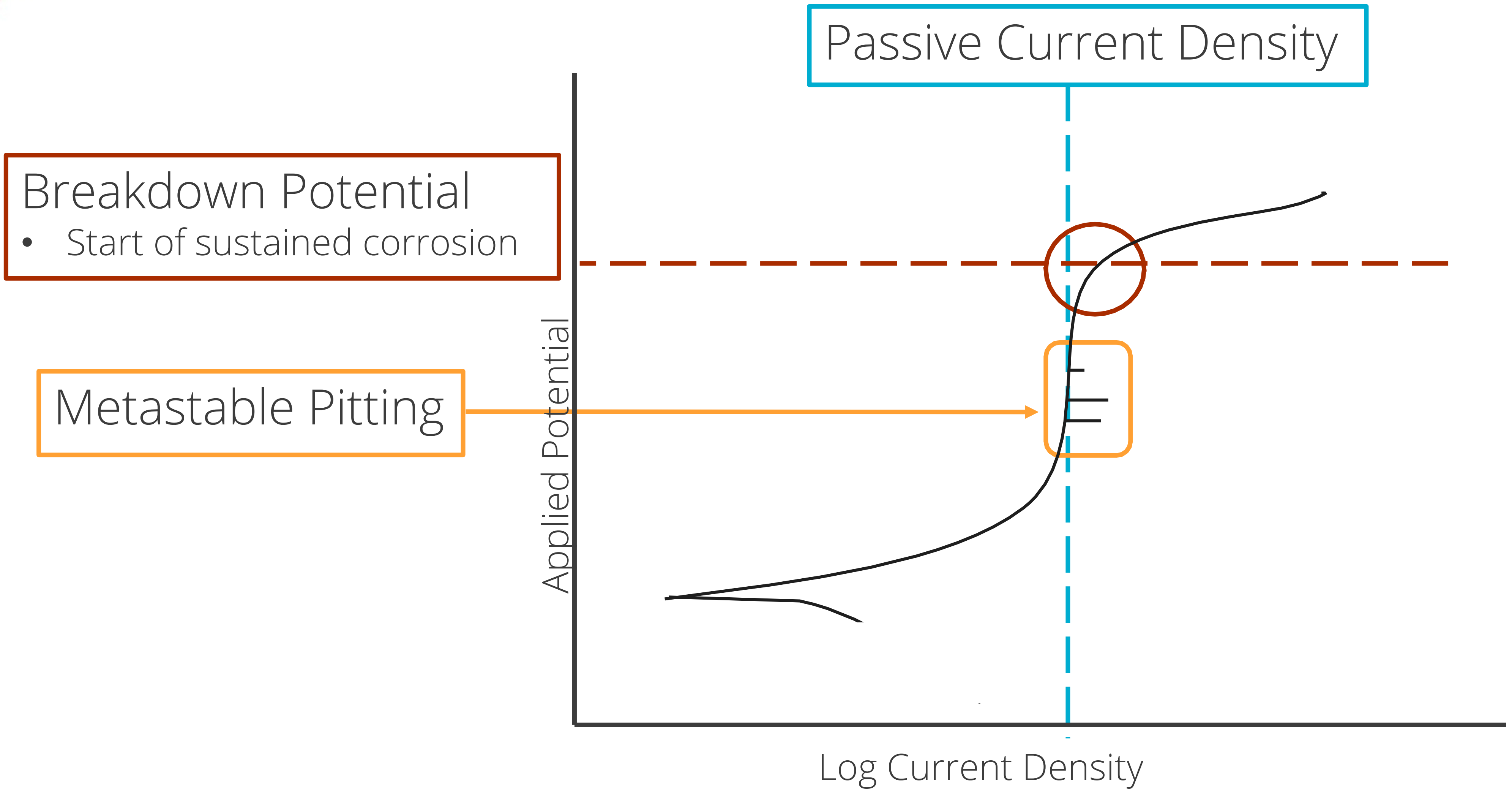
Open Circuit Potential

larger potential difference between cold spray and substrate

higher concern for galvanic corrosion



Potentiodynamic Scans



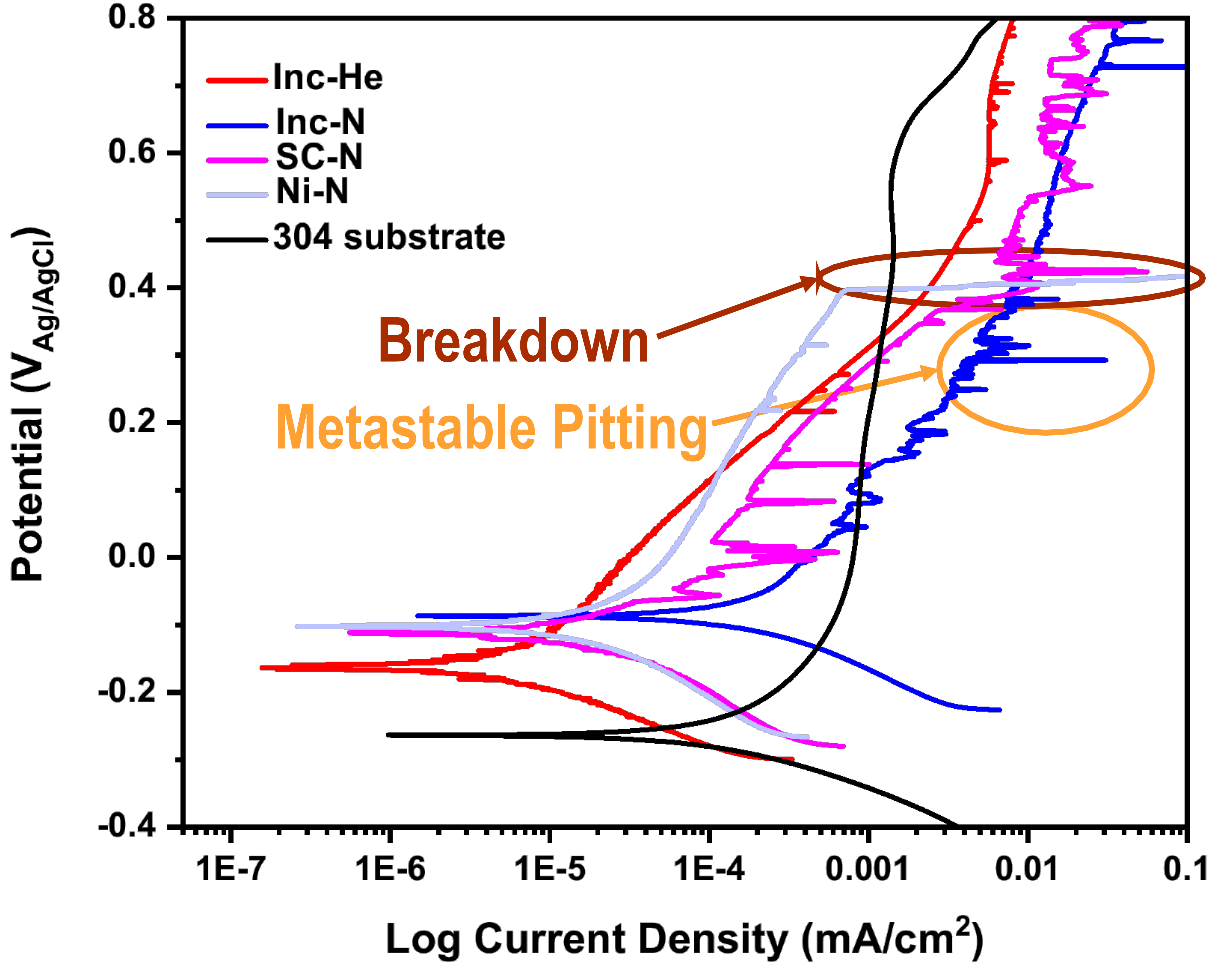
Potentiodynamic Scans: As Sprayed

- Ni breakdown very early in scan
- Metastable pitting

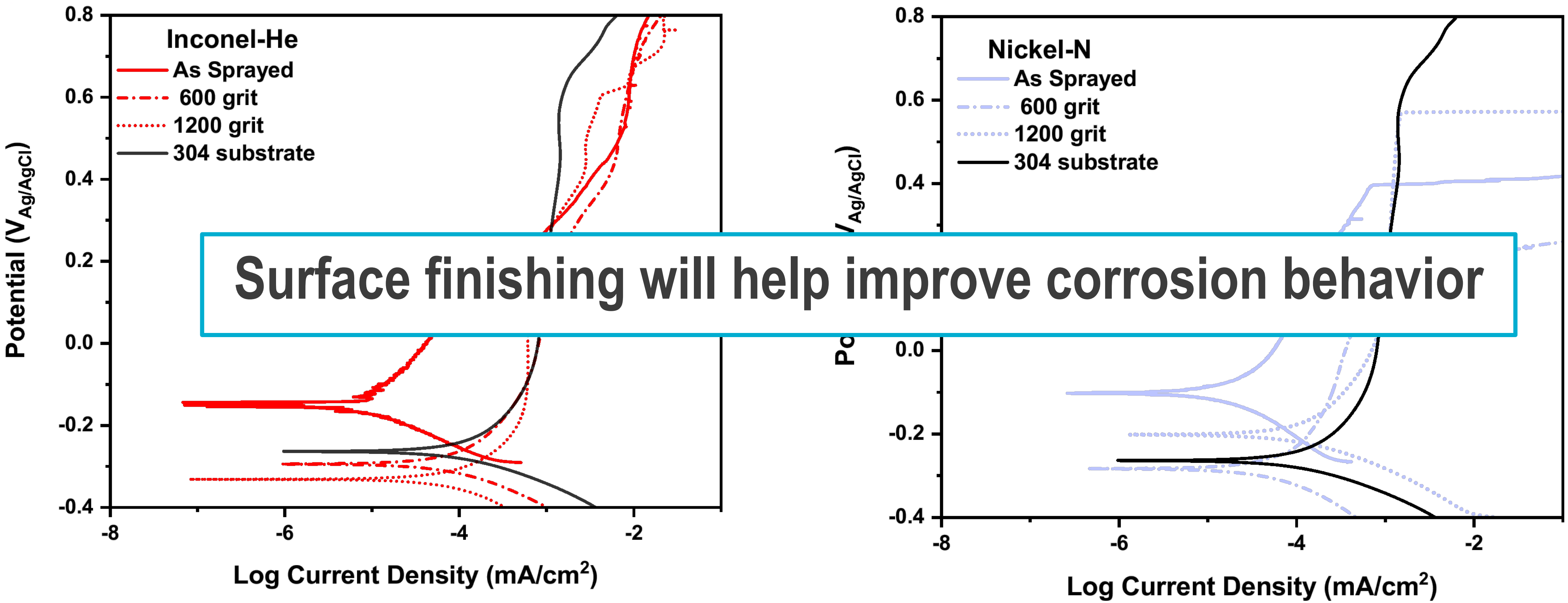
Passive Current Density

- Lower: Ni-N & Inc-He
- Higher: SC-N & Inc-N

Sample	Porosity (%)	S_a (µm)
SC-N	5.51 ± 0.44	16.7 ± 0.5
Inc-He	1.21 ± 0.20	15.7 ± 0.5
Inc-N	5.79 ± 0.18	17.2 ± 0.6
Ni-N	3.78 ± 0.59	18.5 ± 0.6



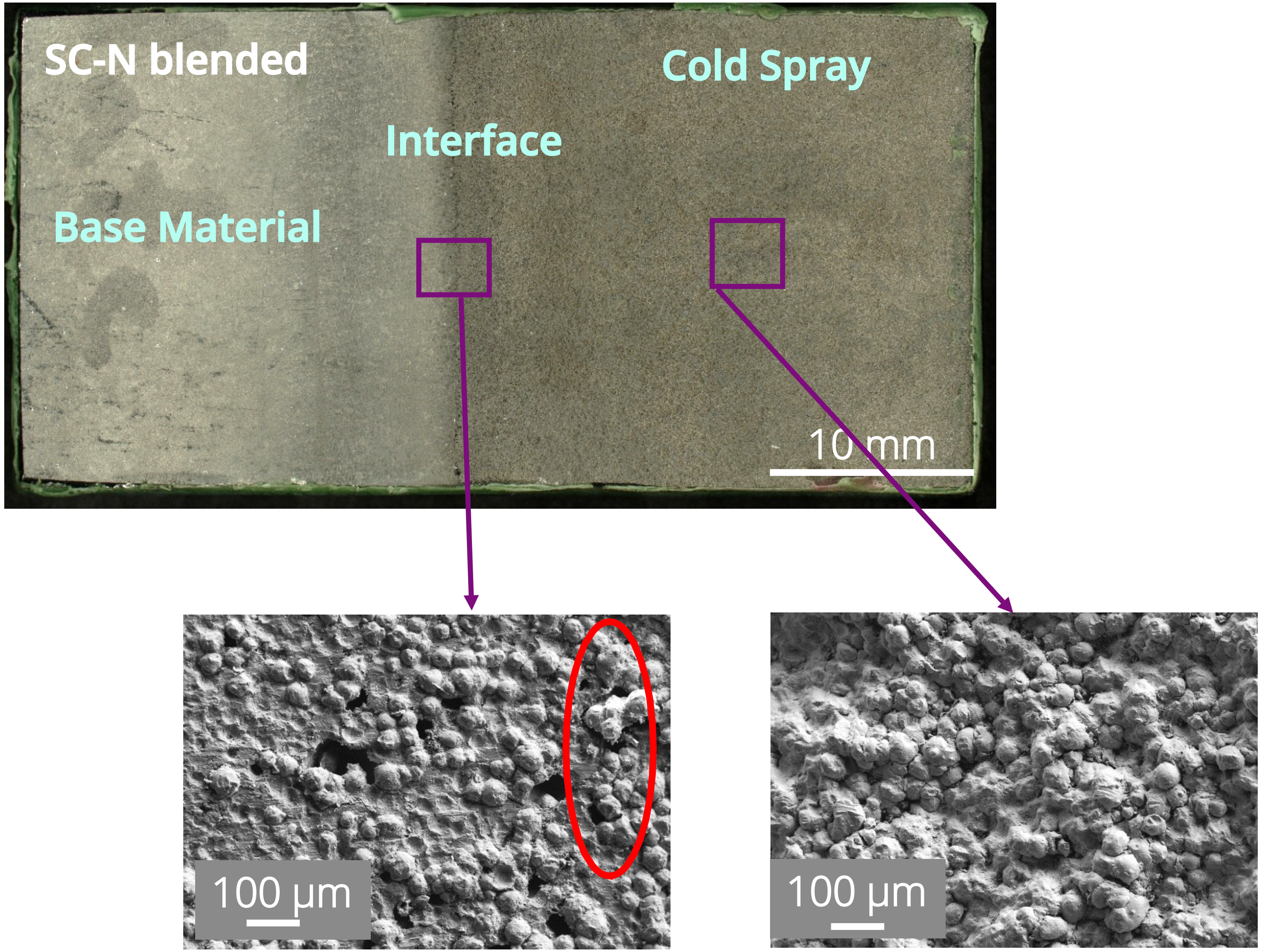
Effects of Polishing



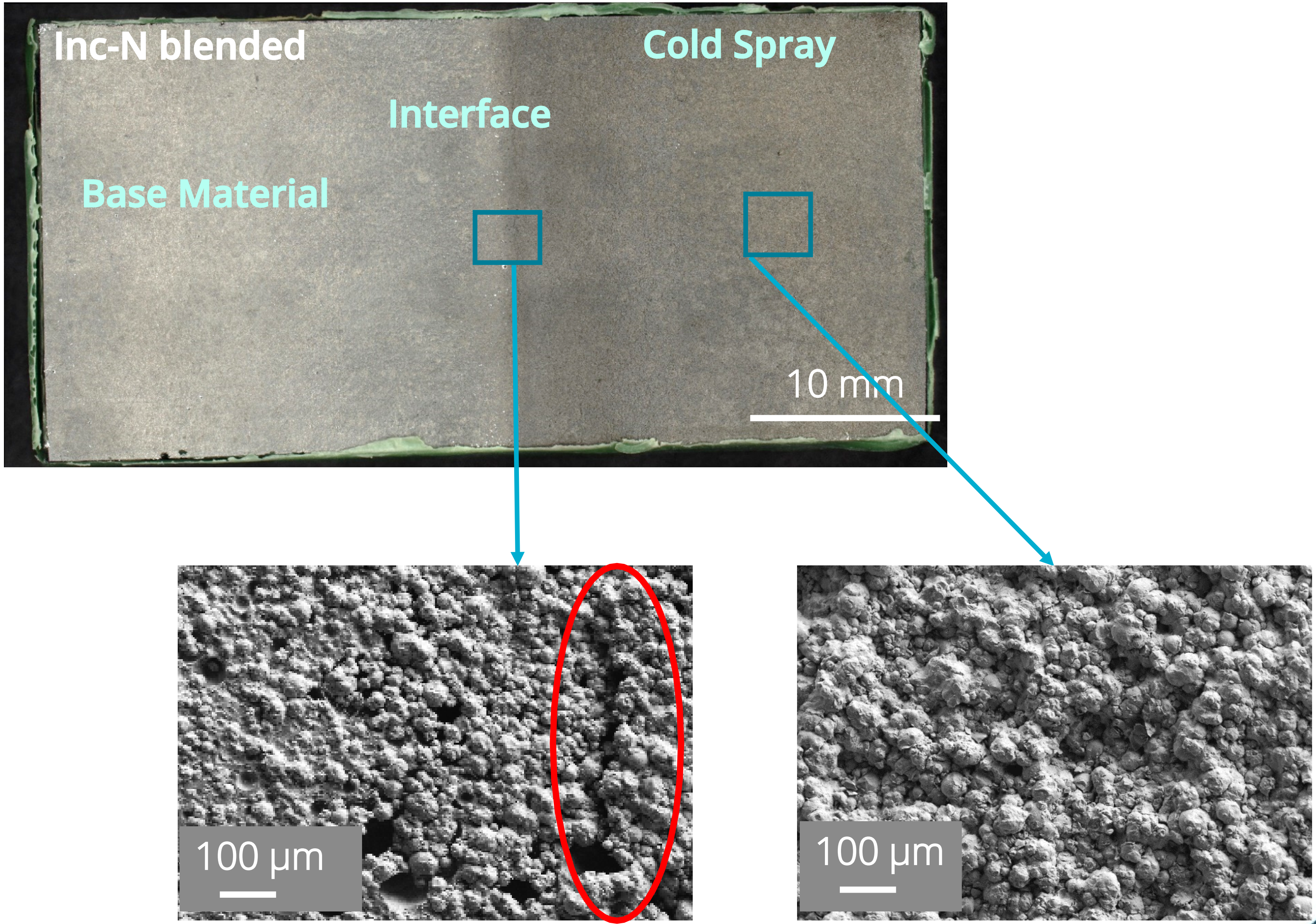
Surface finishing will help improve corrosion behavior

- OCP changes due to polishing (expected)
- Nickel breakdown very early in scan
- Polishing reduces metastable pitting
- Polishing removed native oxide, higher passive current density observed initially

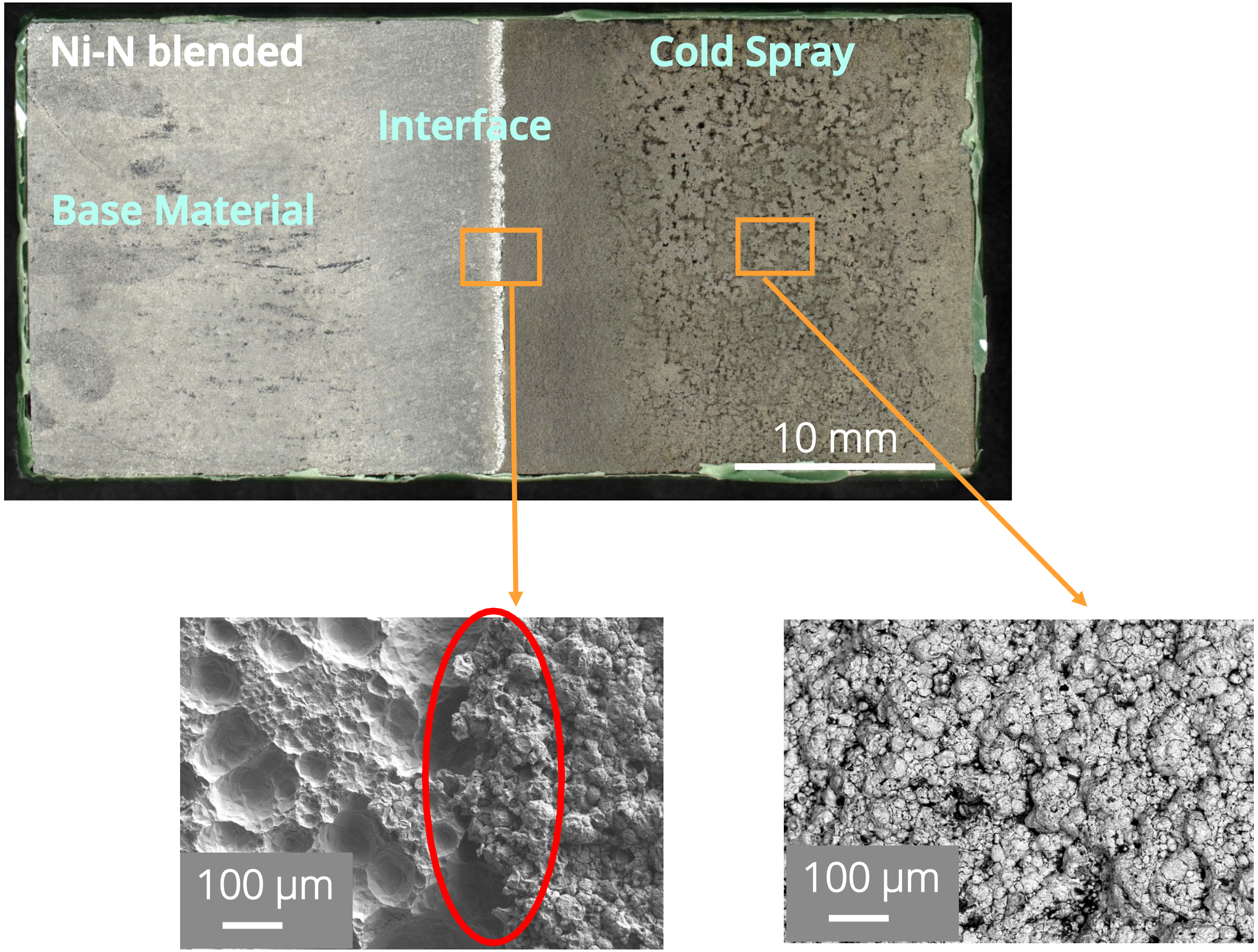
Super C-N Blended



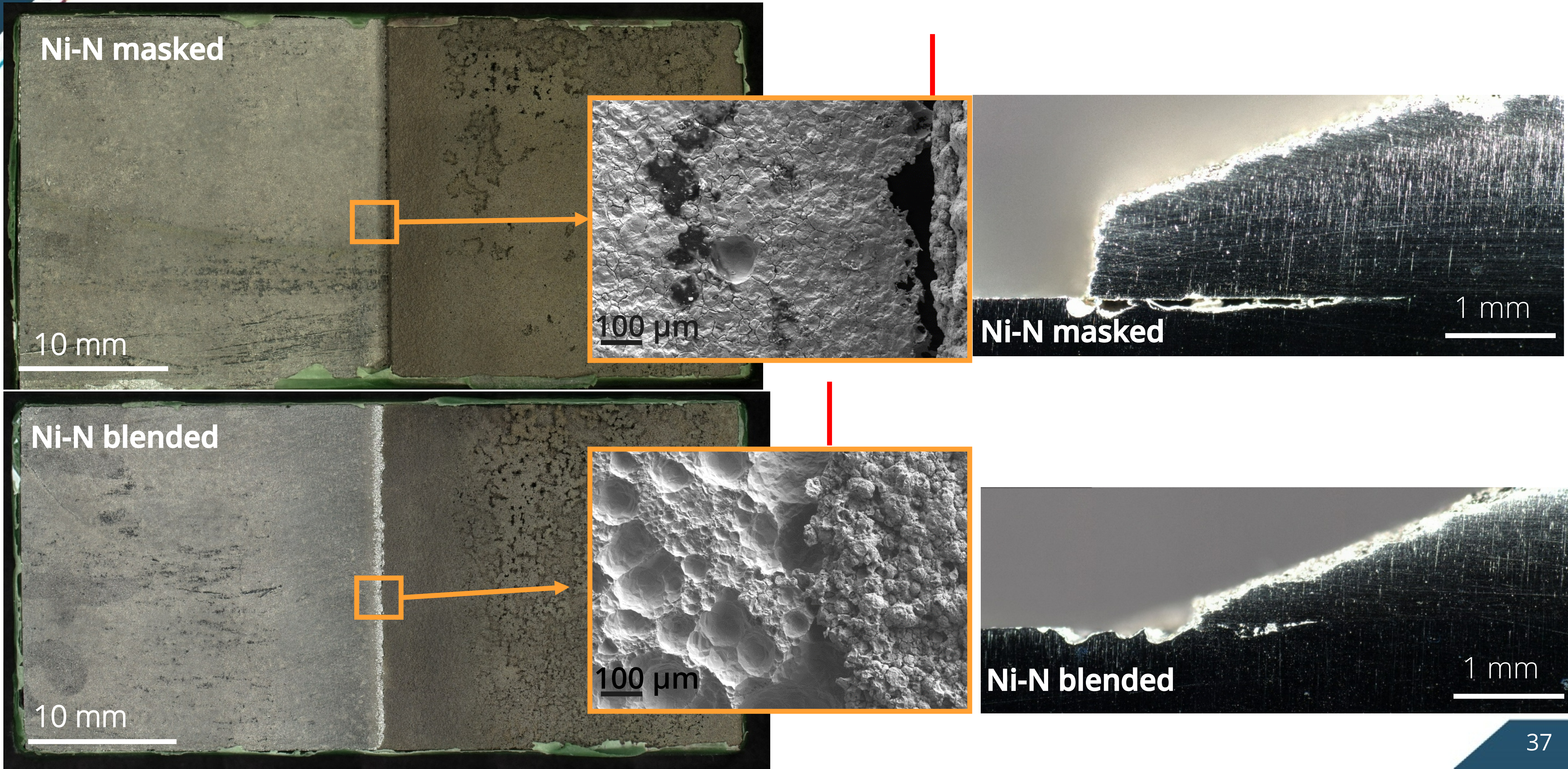
Inconel-N Blended



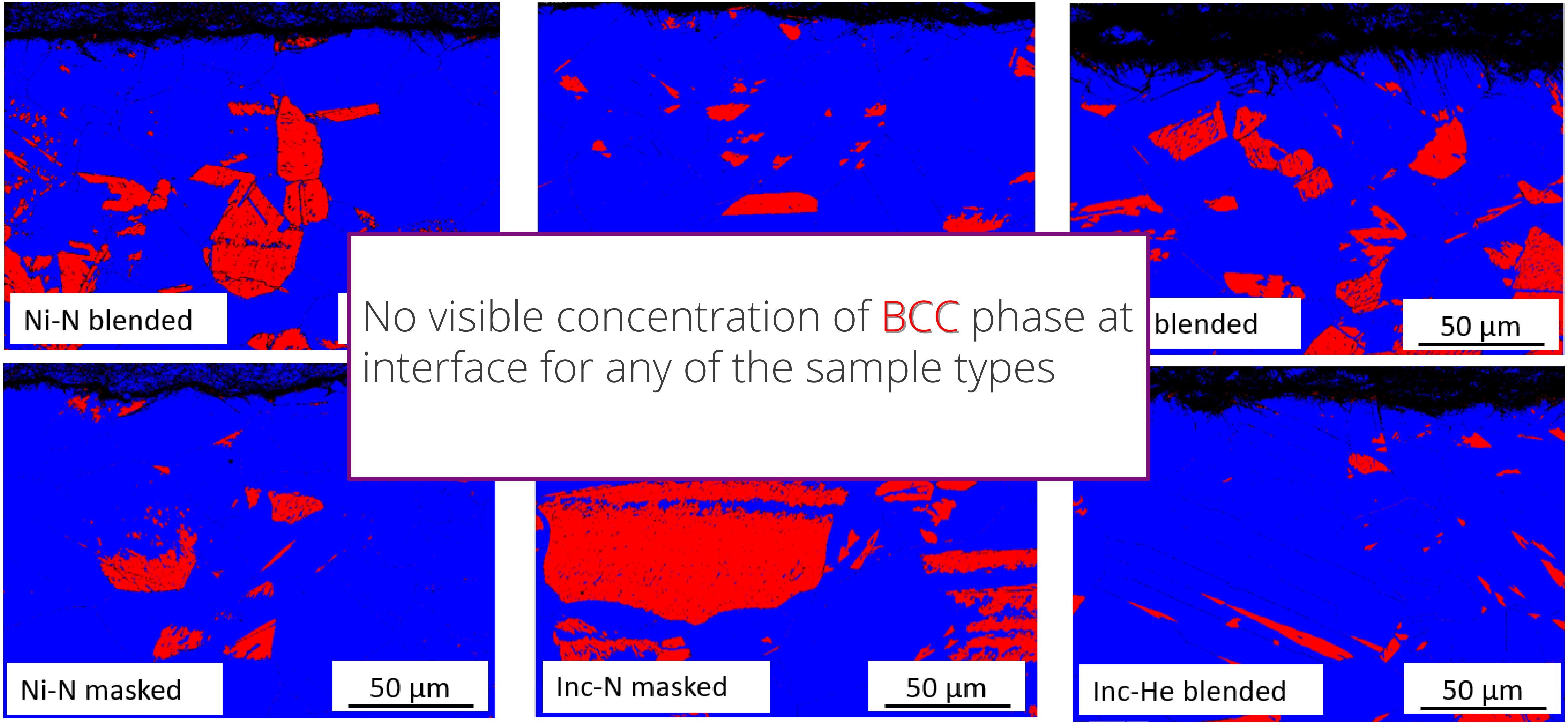
Nickel-N Blended



Masked vs Blended: Ni-N

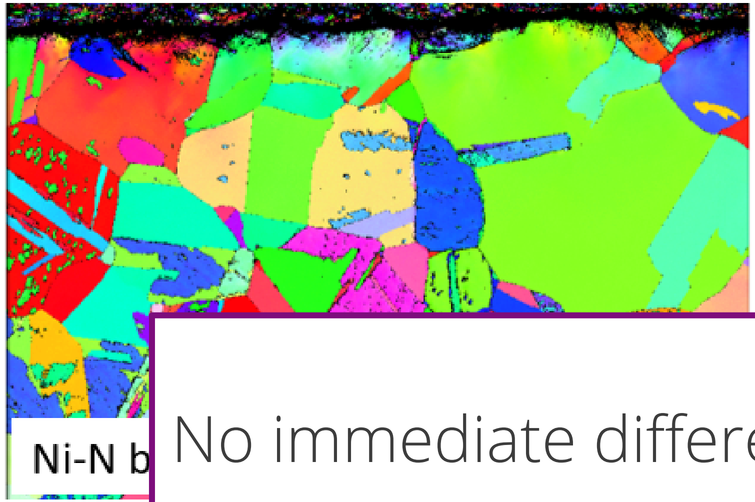


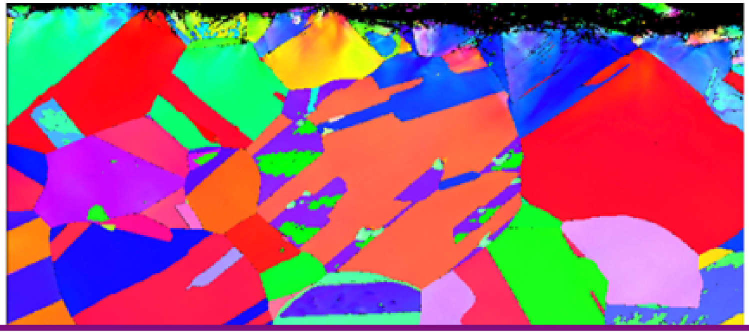
Deformation Layer- Far from Edge

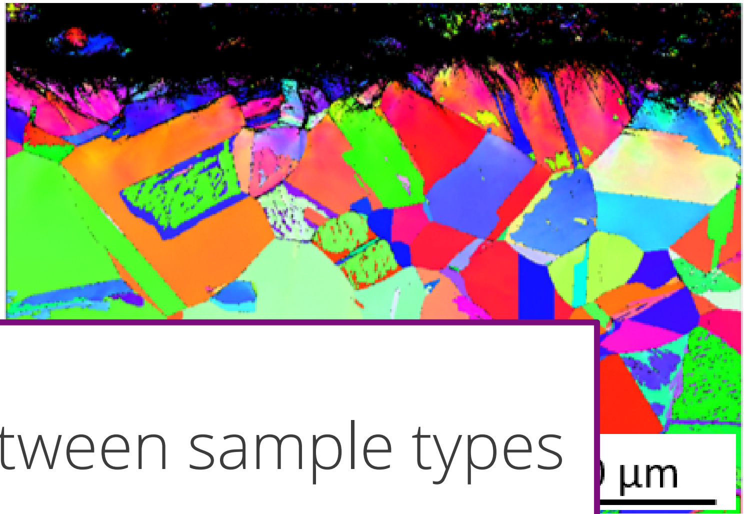


No visible concentration of BCC phase at interface for any of the sample types

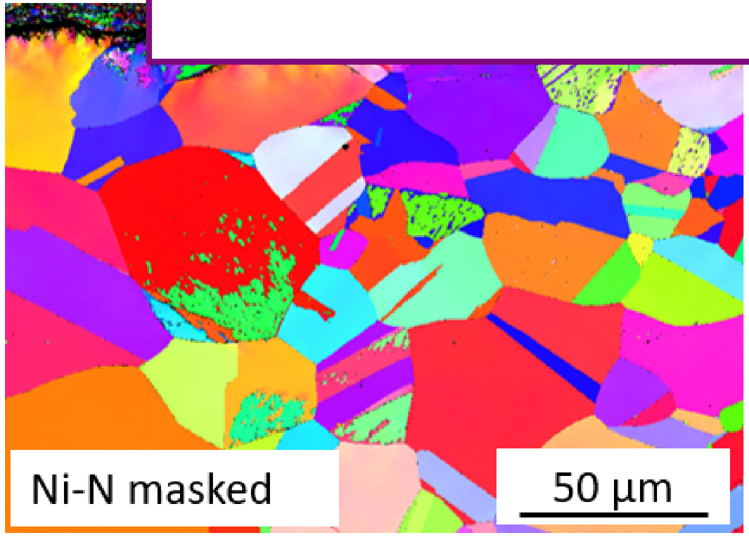
Deformation Layer- Far from Edge

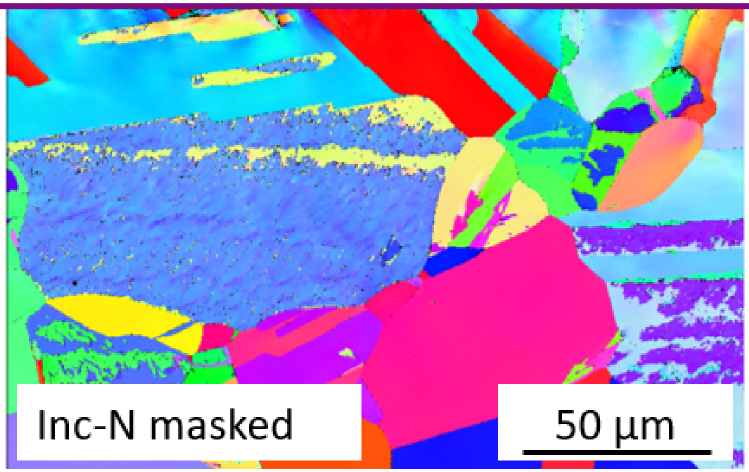


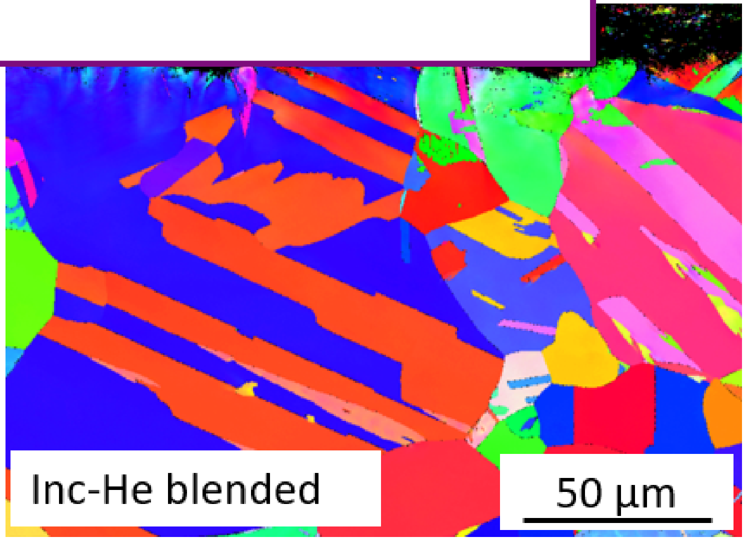




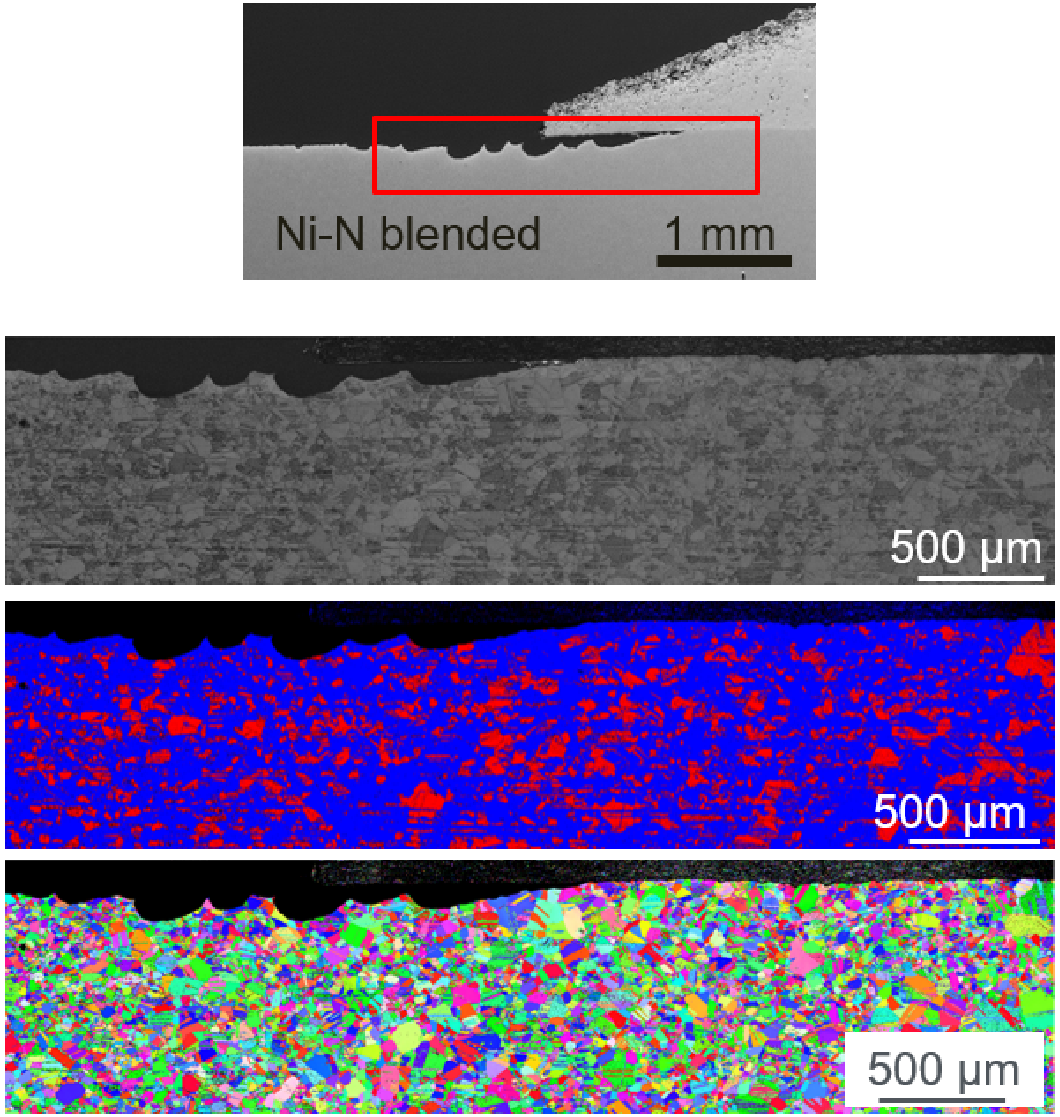
No immediate differences in deformation layer between sample types

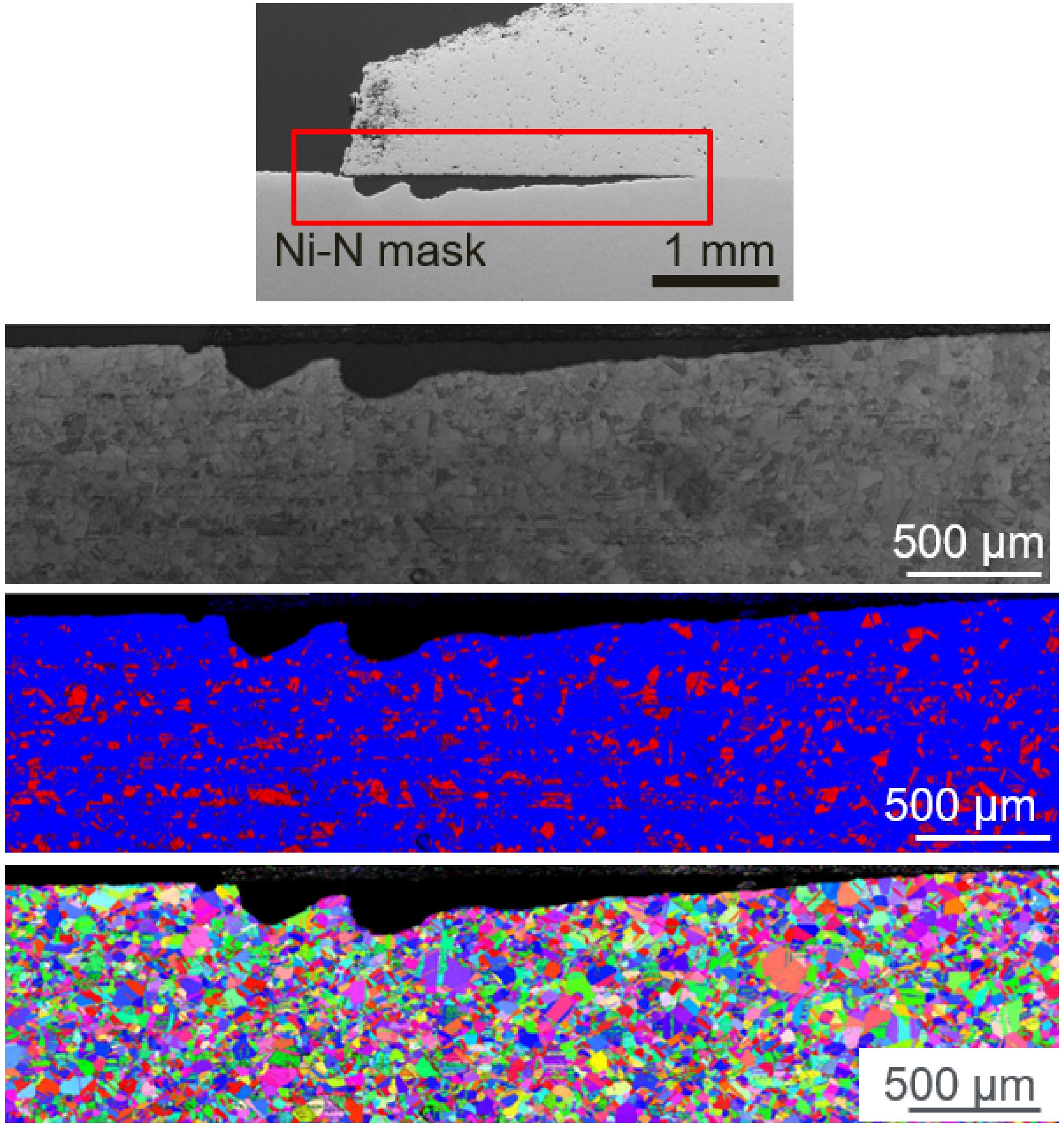




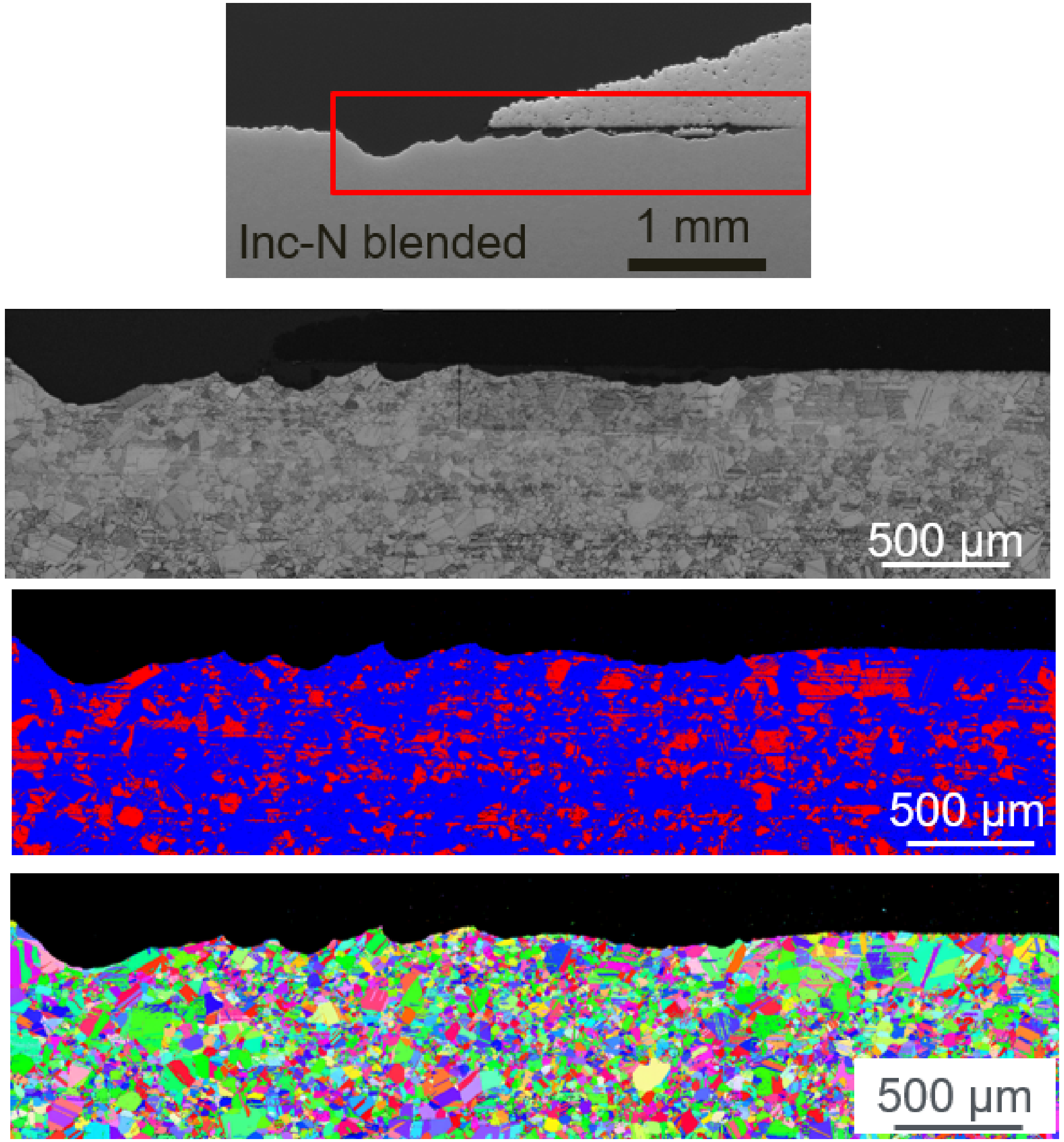


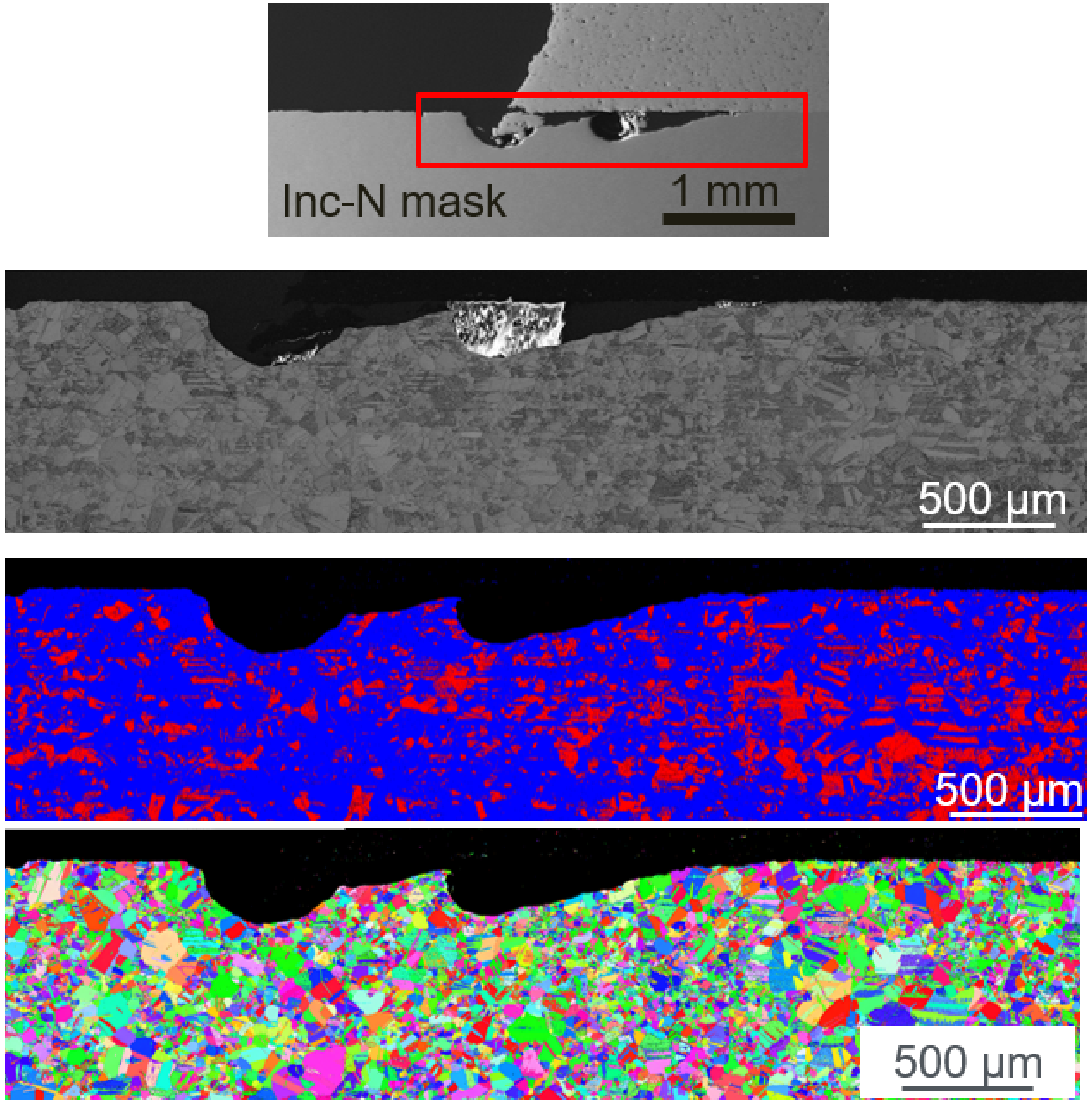
Masked vs Blended: Ni-N



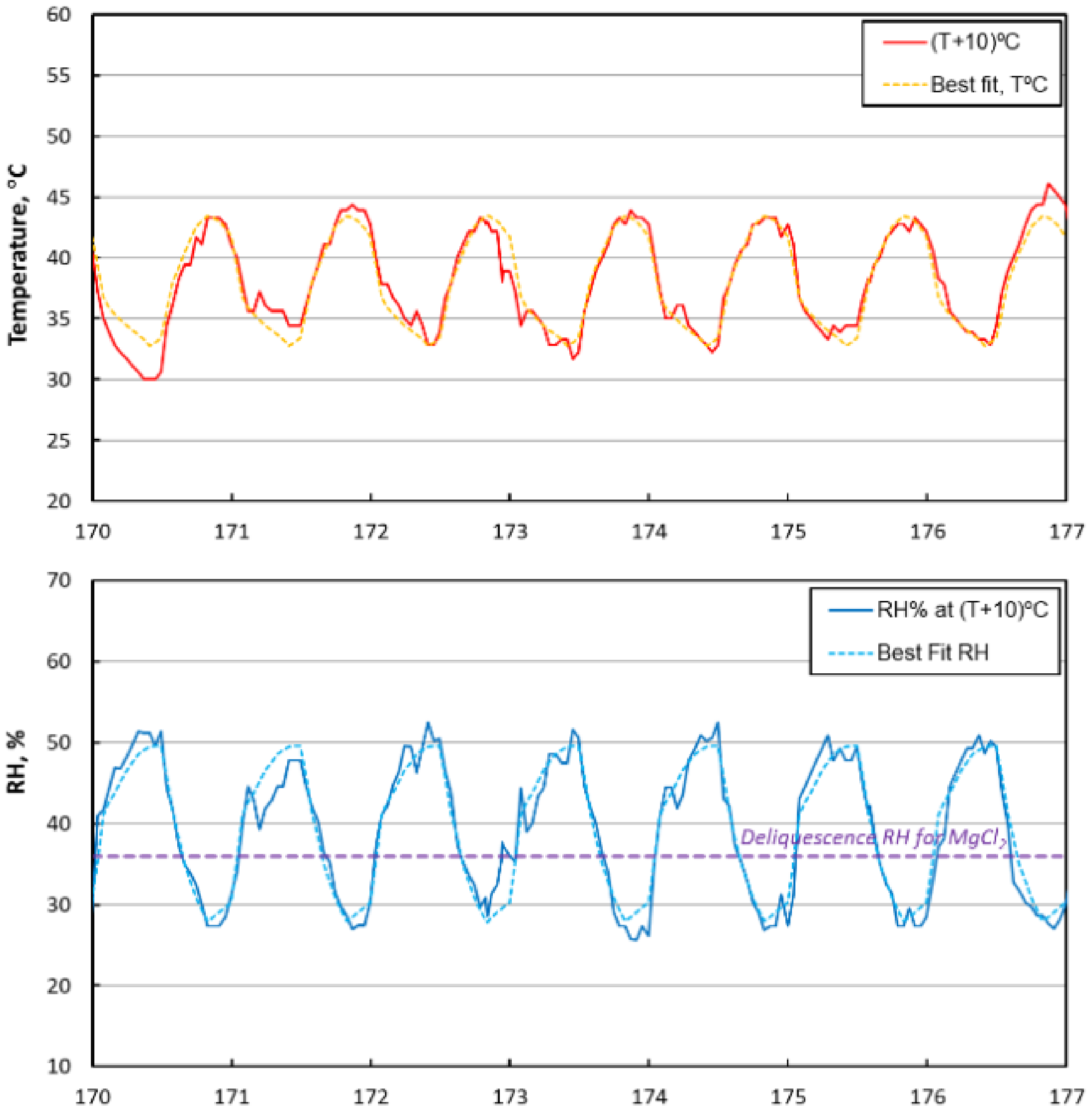


Masked vs Blended: Inc-N





Diurnal Cycle



Hour	Temperature, °C	RH, %
2	41.68	30.33
4	36.45	41.68
6	35.27	43.92
8	34.35	46.69
10	33.69	48.68
12	32.74	49.54
14	33.44	49.57
16	38.24	40.98
18	40.55	34.62
20	42.69	30.62
22	43.51	27.82
24	42.97	29.15

Table 2
Composition of Brines Used for Experimental Testing.

Brine RH	Species concentration (molality)									
	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	Br^-	F^-	SO_4^{2-}	BO_3^{3-}	HCO_3^-
Seawater	0.486	0.010	0.055	0.011	0.567	0.001	0.0001	0.029	0.0004	0.002
90%	2.337	0.050	0.266	0.031	2.723	0.004	–	0.126	0.002	0.001
78%	4.506	0.096	0.513	0.015	5.250	0.008	–	0.196	0.004	0.005
73.4%	5.224	0.111	0.597	0.011	6.108	0.009	–	0.210	0.005	0.010
66%	2.046	0.166	2.700	0.004	6.395	0.047	–	0.568	0.024	0.029
56%	0.718	0.144	3.903	0.003	7.939	0.077	–	0.289	0.040	0.058
38%	0.144	0.032	5.487	0.003	10.602	0.180	–	0.059	0.093[a]	0.195[a]

[a] Concentration of bicarbonate may be significantly over estimated and magnesium carbonate was filtered prior to use thus changing the actual concentration of these components.

Bryan, C. R. et al. "Physical and chemical properties of sea salt deliquescent brines as a function of temperature and relative humidity." *Science of The Total Environment* (2022).