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

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INTRODUCTION

Since its debut in the 1980's, metal additive manufacturing continues to be an expanding area of research. Many techniques have been, and continue to be, developed. One such technique is cold spray. Cold spray is a technique whereby metal particles are accelerated into a substrate material, propelled by a stream of inert gas at temperatures below the melting temperature of the particles. The inert gas is heated, despite the name of the technique, in order to achieve higher particle velocities. These temperatures are usually under 1100 °C[1]. The metal particles adhere to the substrate through a kinetic deformation process, distinguishing it from other thermal spray and additive manufacturing techniques which rely on melting or sintering. Cold spray also induces compressive residual stresses[2] which are often considered beneficial for preventing stress corrosion cracking[3].

The implementation of materials developed through these techniques has rapidly garnered interest. It has seen application in the medical space for biocompatible implant coatings[4], in the military field for vehicle and aircraft repair[5], and as a corrosion protection and wear resistance coating in nuclear energy[6]. Spent nuclear fuel (SNF) canisters are a promising application space for cold spray.

For dry storage purposes, SNF is placed in stainless steel canisters made of 304 or 316 stainless steel. The canisters are placed in concrete overpacks and passively cooled as ambient air passes through vents in the overpack. These are stored at independent spent fuel storage installations (ISFSI) which are often co-located with the nuclear reactors, many of which are in near-marine environments. As a result, chlorides are deposited from the atmosphere onto the canister surface. As the SNF decays, the canisters cool and can eventually reach temperatures where the relative humidity (RH) at the canister surface is high enough to deliquesce the deposited chlorides[7]. These chloride-rich brines can induce corrosion on the surface of the canister and subsequently leads to the potential for chloride-induced stress corrosion cracking (CISCC). There are presently over 3,000 canisters containing over 86,000 metric tons of SNF distributed across 77 ISFSIs[8]. At this point in time, there is no permanent geological repository for the SNF. Therefore, research and development of corrosion mitigation and repair solutions for the dry storage canisters are of significant interest. One of the considerations for addressing in-service canisters is the possibility that coating the entire canister surface may not be feasible. As a result, a patch-type application needs to be considered.

In a patch-type application, an exposed junction between the substrate and cold sprayed material exists. This elevates the potential for galvanic corrosion and introduces a region of potentially high corrosion susceptibility at the patch edge. The following work focuses on this region. A full-immersion ASTM G48 Method A ferric chloride pitting test was used to examine accelerated corrosion behavior at the patch edge for nickel and nickel-based alloys cold sprayed onto 304 stainless steel. Nickel-based alloys, like Inconel, are considered to have good corrosion resistance and have historically been considered a low galvanic corrosion risk when paired with 304 stainless steel, making it a promising material for application on SNF canisters.

METHODS AND MATERIALS

A substrate material of wrought 304L was used and cold spray layer of either Inconel 625-2, Super C, or commercially pure nickel were deposited; compositions are given in Table I. For most of the samples, Nitrogen was used as the accelerating gas, for one sample Helium was used. Two interface types were explored in this work, blended and masked. In the blended case the cold spray layer tapers down to the substrate and in the masked case the drop off to the substrate is abrupt. Samples of each interface, blended and masked, were tested for Inconel-Nitrogen and nickel-Nitrogen. Super C-Nitrogen and Inconel-Helium are presented only in the blended interface.

Experiments were performed in accordance with ASTM G48 method A, ferric chloride pitting test. The cold spray samples were coated in epoxy on the sides and bottom, as seen in Figure 1, to mimic the surfaces that would be exposed in an actual patch application type scenario. The samples were imaged pre-exposure with a Keyence VHX-7000 digital microscope. Samples were immersed for 72 hours at 22 °C in 6% by mass ferric chloride solution. Post-testing, samples were rinsed with DI water and dried with nitrogen gas. They were imaged post-exposure on a Keyence VHX-7000 before being, cross-sectioned, polished to a 1200 grit finish and re-imaged with the same digital microscope.

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TABLE I. Cold Spray Powder Compositions

	Nickel	Super C	Inconel 625
C	≤0.01%	0.02%	0.002%
Co	-	0.2%	0.09%
Cr	-	23.2%	21.47%
Ni	>99.9%	Balance	Balance
Mo	-	17.7%	17.7%
Mn	-	0.7%	0.7%
P	-	0.002%	0.7%
S	<0.001%	0.004%	0.002%
Si	-	0.5%	0.004%
Fe	≤0.14%	0.6%	0.5%
Al	-	-	0.6%
B	-	0.003%	-
Nb	-	-	-
O	≤0.4%	-	≤0.4%
V	-	0.30%	-
W	-	0.26%	-

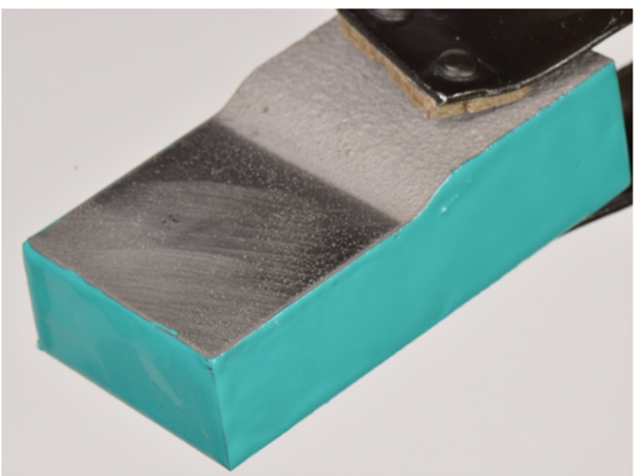

Fig. 1. Cold spray sample with blended interface, coated on sides with epoxy, being held by clamp.

RESULTS

The top-down images post-exposure in the cold spray region for the four different material-gas combinations are shown in Figure 2. Corrosion damage in the cold spray region for the nickel-based alloys was not readily apparent, likely in part due to the inherent surface roughness associated with cold spray. The commercially pure nickel conversely showed quite extensive damage in the cold spray region.

Figure 3 shows optical images of the junction between the cold spray and substrate materials top-down. The nickel sample with a blended interface type sustained substantial damage at the juncture between the cold spray and substrate materials. There were large, overlapping pits that appeared to continue up under the cold spray layer. The nickel sample with the masked interface did not show the same overlapping pits. Instead, the nickel masked interface sample had evidence of crevice formation occurring immediately at the junction between the cold spray and substrate materials.

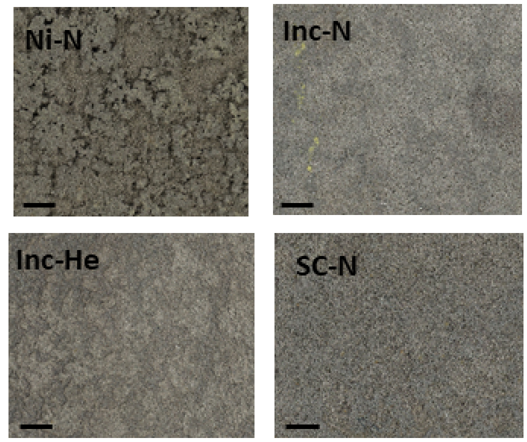

Fig. 2. Optical top-down images in the cold spray regions of samples after ferric chloride testing. Scale bars are 1 mm.

Some large pits can be seen in the interface region, but they are generally not overlapping the crevice. This behavior was consistent with the nickel-based alloy samples with masked interfaces. In all cases, the masked samples showed the same indications the formation of a crevice with large pits near the crevice formed between the cold spray and substrate. The nickel-based alloy samples with blended interfaces had smaller pits, dispersed within the diffuse edge of the blended edge. They also showed evidence of crevice formation, but it was not as wide-mouthed as that of the masked sample.

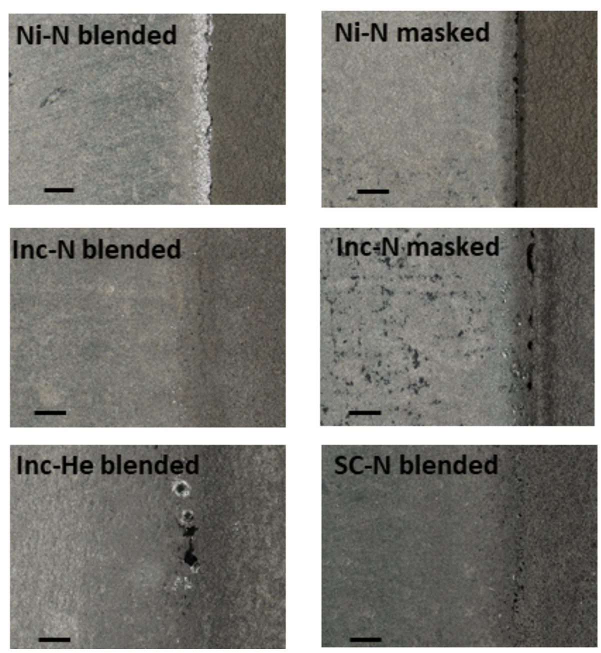

Fig. 3. Optical top-down images at the junction of the cold spray, on the right side of the images, and the substrate material, on the left side of the images, after ferric chloride testing. Scale bars are 1 mm.

The presence of a crevice was more difficult to identify at lower magnifications. There were no immediate differences observed between the Inconel blended interface samples processed with Nitrogen and Helium.

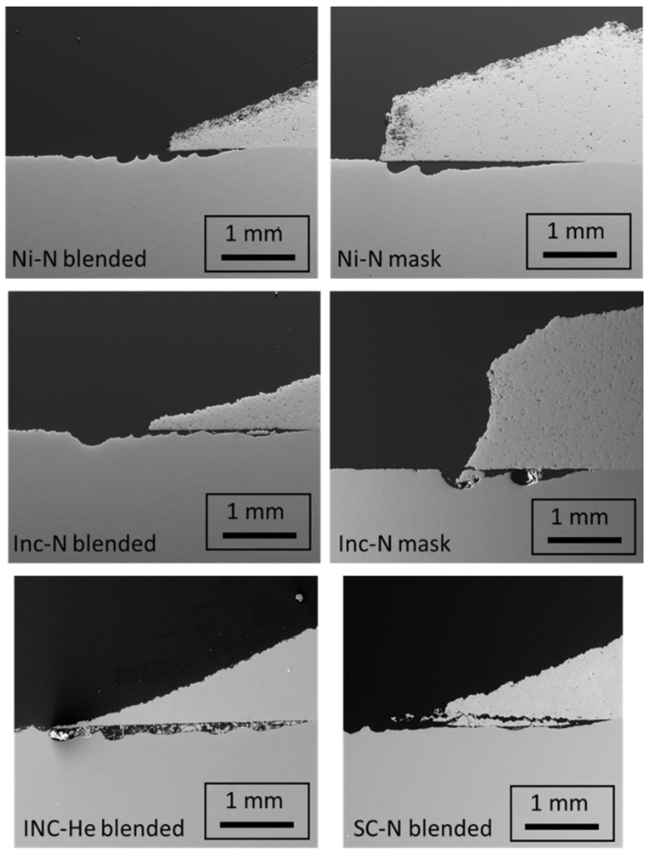

Fig. 4. Scanning electron microscopy micrographs of cross-sections at the cold spray-substrate interface region.

To better assess the extent of the corrosion damage, particularly with regards to the crevice visible in the top-down images, the cross-sections were examined. In cross-section, the damage between the layers becomes readily visible, as seen in Figure 4. As the cross sections are taken at a single location in the samples and therefore cannot divulge the entire story, but important insight can still be gained. Although characteristics of the corrosion damage vary between sample types, pronounced damage can be seen between the cold spray and substrate layers for all of the observed sample types, including the blended interface nickel-based alloy samples for which the crevice was not as readily apparent top-down.

CONCLUSIONS

Only a small set of materials and conditions were used in this study, however all samples showed substantial damage between the cold spray and substrate layers. The cross-sections are only a snapshot of what's happening at one point, but it is notable that corrosion occurs between the cold spray and substrate regardless of material, interface, and processing gas. This is an important factor to consider when selecting a non-destructive examination technique to evaluate the fidelity of the patch after it has been in service and subjected to corrosive environments[9,10].

The material type does affect the corrosion behavior of the sample, both at the interface and in the cold spray region. For the materials tested in this work, the commercially pure nickel suffered damage both in the cold spray and at the interface. The nickel-based alloy samples only showed damage in the interface region and it appeared much less severe than the commercially pure nickel. The differences in corrosion behavior lend credence to the belief that the ASTM G48 Method A ferric chloride pitting test can be used to help compare the coatings' corrosion behavior against one another and rapid comparison of materials to aid in optimization of cold spray composition.

FUTURE WORK

The testing method presented here is an accelerated corrosion test. As a move towards actual application is made, more realistic corrosion environments need to be explored to fully understand the materials degradation behavior. Specifically, atmospheric corrosion tests need to be done. Brine composition, relative humidity, and temperature will likely all play a role in the corrosion behavior. Additionally, other accelerated tests, for the purpose of optimization will also be applied, including, but not limited to, potentiodynamic polarizations in solutions of interest, boiling $MgCl_2$ SCC testing (ASTM G-36), etc. Initial summaries of atmospheric and accelerated testing will be incorporated into presentation at IHLWRM.

ACKNOWLEDGEMENTS

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REFERENCES

1. M. F. SMITH, High Pressure Cold Spray-Principles and Applications, CHARLES M. KAY, J. KARTHIKEYAN, Ed., ASM International (2016).

2. V. LUZIN et al., “Advanced Residual Stress Analysis in Thermal Spray and Cold Spray Processes,” J. Therm. Spray Technol., 29, 1211 (2020).
3. J. TORIBIO, “Residual Stress Effects in Stress-Corrosion Cracking,” J. Mater. Eng., 7, 173 (1998).
4. A. M. VILARDELL et al., “Cold Spray as an Emerging Technology for Biocompatible and Antibacterial Coatings: State of Art,” J. Mat. Sci., 50, 4441 (2015).
5. P. F. LEYMAN & V. K. CHAMPAGNE, “Cold Spray Process Development for the Reclamation of the Apache Helicopter Mast Support,” ARL-TR-4922, Army Research Laboratory (2009).
6. H. YEOM & K. SRIDHARAN, “Cold Spray Technology in Nuclear Energy Applications: A Review of Recent Advances,” Annals of Nuclear Energy, 150, 107835 (2021).
7. C. R. BRYAN et al., “Physical and Chemical Properties of Sea Salt Deliquescent Brines as a Function of Temperature and Relative Humidity,” Sci. Tot. Env., 824, 154462 (2022).
8. “StoreFUEL and Decommissioning Report,” 24, Roswell, GA, UxC StoreFUEL (2022).
9. S. W. GLASS et al., “Cold Spray NDE for Porosity and Other Process Anomalies,” AIP Conf. Proc., 1949, 020010 (2018).
10. D. LEVESQUE, et al., “Laser-Ultrasonic Inspection of Cold Spray Additive Manufacturing Components,” AIP Conf. Proc., 2102, 020026 (2019).