Investigation of Cold Spray as a Dry Storage Canister Repair and Mitigation Tool

Spent Fuel and Waste Disposition

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SUMMARY

The purpose of this work is to investigate the use of cold spray for repair and mitigation of chloride-induced stress corrosion cracking (CISCC) in dry cask storage system (DCSS) canisters to ensure their integrity far beyond their original license period. This report provides a viability analysis of cold spray for repair and mitigation of CISCC in DCSS canisters to extend canister life.

Literature reports that the following three conditions must exist for CISCC to occur: tensile stress, corrosive environment, and susceptible material. These conditions must be understood and accounted for while evaluating CISCC susceptibility. Furthermore, non-obvious factors, such as surface conditions and geometric confinement, can dramatically increase the intensity of one or more of the three listed conditions for CISCC.

It is known that areas in and around fusion welds in alloys used for spent nuclear fuel storage canisters have increased CISCC susceptibility relative to the base metal. Cold spray has the potential to extend the life of DCSS canisters by coating damaged or susceptible areas with cold spray corrosion barriers and inducing beneficial compressive residual stresses.

Cold spray should be considered for manufacturing, CISCC mitigation, and repair of canisters. Cold spray has demonstrated the ability to deposit stainless steel (SS), Inconel alloys, and other alloys on SS 304L. The technical basis for cold spray application within the overpack using remote robotic equipment is established. A non-obvious application to the research community is the sealing of crevices. All instances of CISCC in the field nuclear applications reported by the Pressurized Water Reactor Owners Group were associated with crevice corrosion. This can be avoided by cold spraying to seal crevices.

A significant amount of work needs to be done to develop cold spray for DCSS canister repair and mitigation. Coating powder chemistries needs to be selected such that no detrimental galvanic effects occur and CISCC resistance is maximized. Identifying the optimal chemistry for canister protection is an area that demands a significant R&D effort.

Surface roughness/texture effects are expected to affect CISCC initiation. Cold spray parameters and powder preparation can affect surface roughness. Testing should be done to develop an understanding of how surface roughness/texture developed by cold spray affect CISCC initiation.

This project performed preparatory work and is in the process of generating cold spray samples for CISCC testing by collaborative projects at Pacific Northwest National Laboratory and Sandia National Laboratories. This task will identify the effect of process parameters on microstructure and CISCC resistance. Optimized parameters will be used to produce a smaller number of coupons for full testing and evaluation.

Work on fiscal year 2020 (FY20) scope began when funds were authorized in mid-February. Due to this late start date and delays associated with Coronavirus disease 2019 (COVID-19) part of the FY20 work scope will be completed in FY21 with carryover funds. This report presents an analysis of cold spray for canister life extension, a summary of related efforts, relevant technology down selections, work accomplished to date, and future work relative to the use of cold spray for repair and mitigation of CISCC in DCSS canisters.

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ACRONYMS

ANL	Argonne National Laboratory
APS	atmospheric plasma spraying
BRC	Blue Ribbon Commission on America's Nuclear Future
BWR	boiling water reactor
CAPS	controlled atmosphere plasma spraying
CISCC	chloride-induced stress corrosion cracking
CTE	coefficients of thermal expansion
COVID-19	Coronavirus disease 2019
CNC	Computer Numerical Control
CP	Commercially Pure
CPVC	chlorinated polyvinyl chloride
CSC	cesium and strontium capsule
DCPD	direct current potential drop
DCS	dry cask storage
DCSS	dry cask storage system
DoD	U.S. Department of Defense
DOE	U.S. Department of Energy
DPC	dual-purpose canister
DST	double-shell tank
EM	(Office of) Environmental Management
EPRI	Electric Power Research Institute
ESCP	Extended Storage Collaboration Program
FEA	finite element analysis
FY	fiscal year
HAZ	heat-affected zone
HPCS	high pressure cold spray
HPPS	high-pressure plasma spraying
ISFSI	Independent Spent Fuel Storage Installations
IGSCC	intergranular stress corrosion cracking
ISFSI	independent spent fuel storage installation
LPCS	low-pressure cold spray
LPPS	low-pressure plasma spraying
MCSC	Management of Cesium and Strontium Capsule
NDE	non-destructive evaluation
NRC	U.S. Nuclear Regulatory Commission
PNNL	Pacific Northwest National Laboratory
PVD	physical vapor deposition

SBIR	Small Business Innovation Research
SCC	stress corrosion cracking
SEM	scanning electron microscopy
SFWD	Spent Fuel and Waste Disposition
SNL	Sandia National Laboratories
SONGS	San Onofre Nuclear Generating Station
SPP	Solid Phase Processing
SSs	Stainless Steels
UWM	University of Wisconsin-Madison
VPS	vacuum plasma spraying

INVESTIGATION OF COLD SPRAY AS A DRY STORAGE CANISTER REPAIR AND MITIGATION TOOL

1 INTRODUCTION

The lack of a permanent spent nuclear fuel repository means utilities are faced with extending licenses for dry cask storage systems (DCSS) canisters far beyond their original 20-year license period. In fact, license extension has already been granted at selected sites for up to 40 years with an expectation of further extension of 40 years more (NRC 2012). DCSS canisters are typically made of austenitic stainless steels (SSs) cylinders that are closed using fusion welds to provide leak-tight containment of the spent fuel. With extension beyond their designed service life, chloride-induced stress corrosion cracking (CISCC) has become a major concern to the long-term integrity of DCSS canisters, especially in the welded region. This is because many DCSS canisters are located near salt-water bodies or other sources of chlorides, where chloride (Cl)-rich deliquescent brines can deposit on canisters creating a susceptible environment for CISCC precursor to form. In addition, high residual tensile stress and susceptible microstructure resulted from the high heat input and melting/subsequent cooling during the fusion welding process can provide susceptible mechanical and material conditions for CISCC initiation and propagation. The purpose of the work reported here is to investigate the use of cold spray for repair and mitigation of CISCC in DCSS canisters to ensure integrity far beyond their original license period.

This report presents an analysis of cold spray for canister life extension, a summary of related efforts, relevant technology down selections, work accomplished, and future efforts relative to the use of cold spray for repair and mitigation of CISCC in DCSS canisters.

1.1 Chloride-Induced Stress Corrosion Cracking

Three conditions must exist for CISCC to occur: tensile stress, corrosive environment, and susceptible material (Figure 1). These conditions must be understood and accounted for when evaluating CISCC susceptibility. Furthermore, non-obvious factors, such as surface conditions and geometric confinement, can significantly affect the intensity of one or more of the three listed conditions for CISCC.

DCSS canisters are made from austenitic SSs that contain 16-18 weight percent of chromium (Cr) in the solid solution for enhanced resistance to corrosion. Arc welding is used at all confinement boundary closure locations. High heat input and melting associated with arc welding cause material on the weld line to melt. The molten material shrinks during solidification thereby creating tensile residual stresses.

The region that melts in between the joint line during fusion welding is called the fusion zone. The region adjacent to the fusion zone that does not melt but is affected by weld heat input is the heat-affected zone (HAZ). During fusion welding of austenitic SSs, high heat input causes Cr-rich carbides to precipitate along grain boundaries leading to Cr depletion at grain boundaries, resulting in susceptibility to intergranular (IG) corrosion. This is referred to as sensitization and form of attack is called intergranular stress corrosion cracking (IGSCC). Stainless steels with low carbon content, denoted by “L”, were developed to mitigate sensitization during arc welding.

To provide a technical basis for extended storage capabilities, the U.S. Department of Energy (DOE) performed a gap analysis (Hanson et al. 2012) identifying that the DCS container welds fabricated using conventional fusion welding processes were sensitive to CISCC. Research work by Sandia National Laboratories (SNL) and others has shown that through-wall tensional residual stresses exist in current DCS containers. When combining these characteristics, canister performance over extended periods of time becomes less certain.

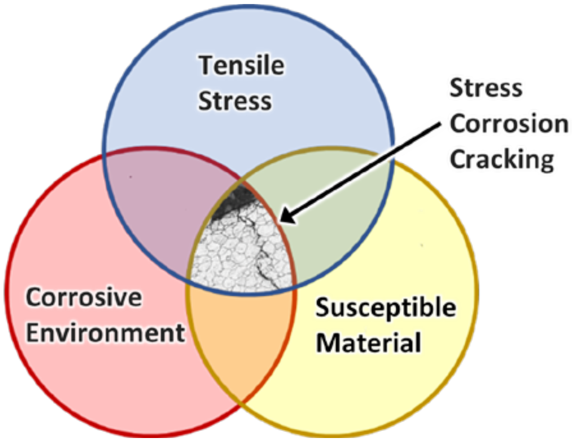

Figure 1. Three conditions required for SCC.

Cold spray is a potential solution to mitigate CISCC in DCSS canisters. No melting occurs and the energy input can be controlled such that detrimental effects to the base metal during these processes are reduced and possibly eliminated. The goal of this work is to develop and demonstrate cold spray with improved CISCC resistance relative to SS 304/304L base metal in and around the processed regions. Cold spray produces a corrosion barrier that separates the susceptible material from a corrosive environment. Furthermore, cold spray can induce compressive residual stresses in the coating and in the substrate material immediately beneath. Canister material directly beneath a cold spray coating is not exposed to the corrosive environment and is in a compressive residual stress condition and CISCC is mitigated.

1.2 Arc Welding

Arc welding uses an electric arc to join metal by melting materials at the interface. This has been a standard technology for joining metals for nearly a century. Currently, arc welding is the standard fabrication and repair technology for the nuclear and many other industries. The DOE (Hanson et al. 2012) and U.S. Nuclear Regulatory Commission (2012) determined residual stresses resulting from fusion welds in austenitic DCSS canisters put the fusion weld areas at higher risk for CISCC than for base metal.

1.2.1 Advantages

Arc welding is currently used for fabrication and repair of steel components for many applications. Some advantages of arc welding relative to canister repair are listed below.

- Manual and automated equipment are commercially available.
- Robotic solutions for repair within overpack are at a high technology readiness level.
- Relevant codes and code cases exist for various arc welding repair processes.

1.2.2 Technical Challenges

- A fundamental physics issue causes reduced CISCC resistance of material in and around arc welded zones.
- Weld shrinkage associated with resolidification produces residual tensile stress in and around the weld making it susceptible to further degradation such as cracking.
- High heat input and melting, and subsequent cooling cause microstructural and microchemical changes in and around welds making them more susceptible to corrosion and SCC.

Arc welding, shown in Figure 2, is considered for temporary and permanent repairs. It is known that alloys used in the existing inventory of spent nuclear fuel storage canisters have reduced CISCC resistance in areas around fusion welds. Ideally, fusion welding used for repair should be combined with a mitigation technique, such as cold spray coating on the weld and the HAZ.

Figure 2. Arc welding of a DCSS canister provided by Fluor.

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2 COLD SPRAY FOR CANISTER LIFE EXTENSION

Cold spray is a technology of interest for repair and mitigation of CISCC in DCSS canisters. Canister life in this report refers to extending canister life through CISCC repair and/or mitigation techniques. This section provides a technical background and discusses the potential application of cold spray relative to canister life extension.

2.1 Process Description

Metal spray coating processes use heated gas to propel metal particles that bond to a substrate material. Thermal spray is a family of metal spray coating processes where particles are fully or partially melted during the process and re-solidified after impacting the substrate. Thermal spray processes include plasma spray, detonation spray, high-velocity oxy fuel spray, and their variants. Because melting and resolidification occur during thermal spray, tensile residual stresses are caused by the shrinking that takes place during resolidification. Oxidation and undesirable chemical reactions also occur due to high heat input and melting. Most thermal spray processes are limited in build thickness to 1 mm or less. In limited cases build thickness can be somewhat thicker. Mechanical interlocking of solidified particles is the bonding mechanism for thermal spray.

Cold spray is a solid phase metal spray process during which no melting occurs. Metal particles are carried by a heated gas stream that softens the metal and propels particles at high velocities. The impact energy is high enough to bond metal particles to the surfaces they impact. Because it is a solid phase process, cold spray avoids oxidation, tensile residual stresses, and other detrimental effects typical of the high heat input and melting associated with thermal spray. Cold spray can produce infinitely thick coatings with beneficial compressive residual stresses.

Some researchers in the thermal spray community describe cold spray as a type of thermal spray. This is incorrect. Melted particles are part of the definition of thermal spray (TWI 2020; VRC 2020). The technical driver for cold spray development and commercialization was the avoidance of issues associated with high heat input, melting, and resolidification that occur during thermal spray processes. For the purpose of this report, cold spray refers to metal spray processes in which no melting occurs and thermal spray refers to metal spray processes in which melting occurs.

Cold spray can produce superior properties relative to thermal spray for materials of interest in spent nuclear fuel storage and transportation applications. Furthermore, oxidation and tensile residual stresses are attributes of thermal spray that impair properties and make the material more susceptible to CISCC. Therefore, thermal spray processes are low priority for evaluation for CISCC repair and mitigation.

High-pressure cold spray (HPCS) is the metal spray process of greatest interest for CISCC repair and mitigation. Figure 3 shows a diagram of HPCS where particles are accelerated to supersonic velocities and impact a substrate. Figure 4 shows nickel deposited using HPCS. During the process, substrate heating is minimal, dimensional stability is maintained, and unwanted thermal effects (HAZ, thermal stresses, dilution layer formation, etc.) are avoided. HPCS systems operate at pressures typically ranging from 300 to 1,000 PSI (VRC 2020) and typically produce particle velocities ranging from 800 to 1400 m/s (Moridi et al. 2014). High velocity enables high kinetic energy, which is required to create high plastic deformation and shearing at particle surfaces. This results in dynamic recrystallization and metallurgical bonding at interparticle boundaries. Particles are held to each other and to the substrate by both mechanical interlocking and metallurgical bonding.

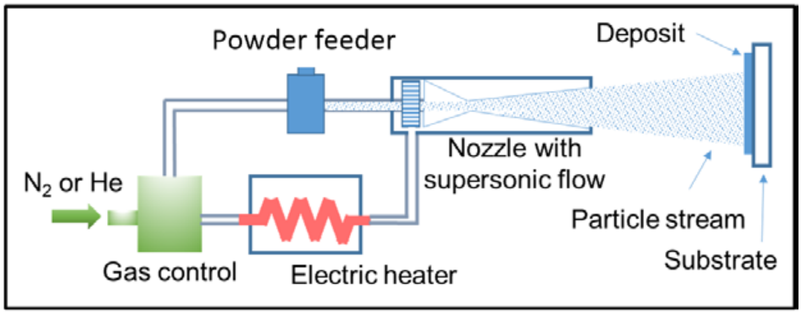

Figure 3. High-pressure cold spray process diagram.

Figure 4. High-pressure cold spray coating of commercially pure nickel sprayed (left side) at PNNL.

Low-pressure cold spray (LPCS) is of lesser interest because it fails to propel particles fast enough to achieve the kinetic energy needed for high-quality cold spray deposition of alloys with high melting temperatures. LPCS systems operate at 300 PSI and lower (VRC 2020). They typically produce particle velocities ranging from 300 to 600 m/s (Moridi et al. 2014). Reduced kinetic energy associated with LPCS means less plastic deformation, less interlocking, and no or dramatically reduced metallurgical bonding in materials with high melting points. Reduced kinetic energy means deteriorated mechanical properties relative to HPCS. LPCS systems are not recommended for high-quality cold spray of steels, Inconel, and other high strength/melt temperature materials.

Kinetic metallization, pulsed gas dynamic spraying, vacuum cold spray, and warm spray are cold spray variants. These variants have not demonstrated the ability to match properties that can be achieved using HPCS for materials of interest for spent nuclear fuel canister repair and mitigation.

The rest of this section focuses on HPCS because it has advantages and improved properties relative to competing metal spray techniques.

2.2 Corrosion Protection Mechanism

Three conditions must exist for CISCC to occur: tensile stress, corrosive environment, and susceptible material (Figure 1). These conditions must be understood and accounted for when evaluating CISCC susceptibility. Furthermore, non-obvious factors, such as surface conditions and geometric confinement, can significantly affect the intensity of one or more of the three listed conditions for CISCC.

Cold spray provides a corrosion barrier coating that has the added benefit of generating compressive residual stresses in the coating and the material immediately beneath the coating. This effectively removes two of the three conditions required for CISCC. For the sensitized material around the welds beneath the coating, the two removed conditions are exposure to corrosive environment and tensile stresses. For the cold spray coating, the two removed conditions are tensile stresses and susceptible material.

2.3 Properties

High CISCC resistance of nickel and nickel-based alloys has been reported by industry and academia (Alloys 2020; Haynes 2020). Various metal vendors report CISCC data correlating CISCC resistance to the fraction of nickel contained in each alloy. Discussion of cold spray properties in the paragraphs below is relative to common canister materials (SS 316 and SS 304) and nickel-based alloys.

In 2012, Westinghouse reported results from exploratory testing done to evaluate the use of HPCS for prevention of pressurized water reactor primary water SCC (Lareau et al. 2012). Cold spray coating of commercially pure (CP) nickel was applied to Inconel Alloy 600 substrates. Cyclic fatigue was evaluated with four-point bend testing; 50,000 cycles with 22.5 ± 21 ksi tensile stress loading was applied. Non-destructive evaluation (NDE) and scanning electron microscopy (SEM) work showed no cracking or debonding. Thermal cycling was done by heating coated samples to 400°C and plunging them into water. After 100 cycles no indications of cracking or debonding were found using NDE techniques and SEM. Impact testing was done with a round-nosed weight with 10J of energy. No cracking or spalling was observed. Vickers hardness testing performed on polished cross sections of coating showed remarkable consistency in hardness (~250 Vickers hardness number).

Adhesion testing was done using epoxy-based pull tests. The epoxy failed at ~10 ksi. This means the cold sprayed CP nickel has an adhesion strength of at least 10 ksi.

Doped steam SCC testing was done at 750°F, 5–13 psia H_2, 80 ppm of F-, Cl- and SO_4^{2-}. CP nickel coated and uncoated strain-hardened $^1/_8$” Inconel Alloy 600 and ¼” thick Inconel Alloy-182 clad bend bars were tested. Specimens were put into a four-point bend fixture and samples of each coating/thickness combination were tested at 70, 75, and 80 ksi tensile stress. All uncoated samples failed within 200 hours. Testing stopped after 800 hours for the ¼” samples and after 1,000 hours for the $^1/_8$” samples. No cracking was observed in the coated specimens. Although these tests are not representative of DCSS canisters, these tests provide evidence of the potential benefits of cold spray coatings.

For alloys that have high melting points, best properties are achieved under the following conditions:

1. An HPCS system is used.
2. Helium is used as the carrier gas.
3. Surface preparation is done correctly.
4. The correct material is selected for the application.
5. Powder is processed correctly.
    a. Sieving powder to remove fines.
    b. Drying powder.

For alloys with high melting points, much of the cold spray work that has been done is for coating of jet turbine blades and natural gas power generation turbine blades. These blades operate at temperatures of 1,000°C and higher while rotating at speeds greater than 10,000 rotations per minute (RPM) in corrosive combustion environments. Competing General Electric and Siemens patents exist in the cold spray patent space related to the coating of gas turbine blades. Inconel 718 is an alloy of interest for gas turbine blades. Some reported values for the properties of cold sprayed Inconel 718 are shown in Table 1 below. These numbers come from several universities that used nitrogen as the carrier gas for cold sprayed coupons. Using helium as the carrier gas produces superior properties in cold spray coatings. Information about commercial cold spray coatings for natural gas industries are typically trade secrets and likely have properties superior to what is reported in Table 1 below.

Cold spray coatings that feature high hardness and strength are being developed to replace electroplated chrome and nickel for U.S. Department of Defense (DoD) combat systems. Properties for one such cold spray coating, Ni and CrC-NiCr blend, are listed in Table 1 below. These DoD cold spray coatings are designed to produce greater resistance to corrosion and wear resistance greater than electroplated chrome coatings.

Table 1. Cold spray property values from various sources.

Coating Material	Substrate Material	Carrier Gas	Bond Strength (KSI)	Hardness (HV)	Porosity (%)	Residual Stress (psi)	Ultimate Strength (KSI)	Reference
SS 304	SS 304	N_2/He (25/75)	>~12[(a)]	450	0.07	-50.8 to -65	-	(Yeom et al. 2020)
Inconel 718	SS 316	-	-	507	0.25	-	67	(Luo et al. 2018)
Inconel 718 PWHT	SS 316	N_2	-	~410	<0.5	-	158	(Luo et al. 2018)
Inconel 718	Inconel 718	N_2	>~12[(a)]	-	<2	-29,008 to -58,015	-	(Fiebig et al. 2020)
Ni, CrC-NiCr blend	-	He	38	400-500	<0.5	-	-	(Nardi et al. 2019)

(a) Denotes that epoxy-based adhesion tests are performed and the epoxy failed before coating.

It should be noted that cold spray is an approved repair processes for dozens of DoD applications. Cold spray repaired components have been in service on various military aircraft and naval vessels for nearly a decade (Champagne 2018). Cold spray is also used for repair of commercial aircraft components. Companies such as Detroit Diesel use cold spray to repair/rebuild commercial freight engines. These successful applications demonstrate long term performance when exposed to corrosive environments and other degradation mechanisms. While these conditions are not representative of DCSS conditions, they set a precedent for long term performance in corrosive environments.

2.3.1 Adhesion

All reported values in Table 1 show that adhesion strength is greater than ~12 ksi, which is the pressure at which the epoxy used for adhesion testing fails. Adhesion values for all material of interest for HPCS CISCC repair and mitigation of spent nuclear fuel canisters will have adhesion values far greater than 12 ksi. Triple lug shear testing, described in MIL-J-24445A (Specification 1977), can be used to get adhesion values not limited to epoxy strength. Triple lug shear testing is far more expensive than epoxy-based adhesion testing and often is not done. Values obtained using triple lug shear show that adhesion values for cold sprayed coatings can be more than triple what can be measured with epoxy-based adhesion tests.

2.3.2 Permeability

Cold spray has no interconnected porosity. When best practices are followed, cold spray coatings for materials of interest should have porosity values less than 1%. Cold spray is used for corrosion barriers in the automotive, defense, and aerospace industries. Canada plans to coat portions of spent fuel canisters with cold sprayed copper for final disposal because they assume it will provide a nonpermeable corrosion barrier over a million years (Hall and Keech 2017).

2.3.3 Mechanical properties

The mechanical properties reported in Table 1 suggest HPCS has excellent mechanical properties, superior to other coating techniques. It is important to understand that some mechanical properties of a part are a combined effect of the substrate and coating and can be affected by coating thickness. These effects can be easily explored using finite element analysis (FEA) and laboratory mechanical testing. Cold spray coatings are less ductile than base metal, but much stronger than non-metallic coatings. High hardness and compressive strength mean that cold spray coatings should do well if a canister is dragged across rails or scrapes the sides of the overpack.

2.3.4 Thermal Concerns

To our knowledge, no data for thermal cycling of cold spray coatings on austenitic SS exists. Thermal cycling of cold sprayed CP nickel on Inconel Alloy 600 was done by heating coated samples to 400°C and plunging them into water. After 100 cycles no indications of cracking or debonding were found using NDE techniques and SEM (Lareau et al. 2012). Cold spray of alloy 718 is used for jet turbine blades that operate at temperatures above 1,000°C and cool when engines are turned off. In these results, coating and substrate materials are nickel-based and have similar coefficients of thermal expansion (CTEs) and are exposed to thermal cycles far more extreme than what could be experienced in a DCSS environment. Therefore, we can assume that thermal cycling for cold spray coatings with similar CTEs to substrate material will be an issue of little concern. Due to relatively small changes in temperatures experienced in DCSS canisters, relative to other cold spray applications, mismatches in CTE's between coating and canisters may be acceptable. Thermal cycling tests should be done for coating-substrate combinations with significantly different CTEs and considered for all coatings.

HPCS using best practices and appropriately selected materials should have no thermal concerns for DCSS canister applications.

For field repair, seasonal variation in temperature should have no significant effect on coating quality because the substrate is heated by the heated gas stream. Heated gas is run through the nozzle, heating the system and substrate prior to injecting powder to ensure steady-state conditions are achieved prior start of material deposition.

2.3.5 Corrosion Resistance

Canada plans to coat portions of spent fuel canisters with cold sprayed copper for final disposal because they assume it will provide a nonpermeable corrosion barrier that lasts more than a million years (Hall and Keech 2017) in a geological disposal site. For DCSS canisters, made of austenitic SS, materials with high CISCC resistance can be applied to sensitized regions in and around welds. This means that base metal, not affected by fabrication welds, becomes the weak link for CISCC susceptibility. If the proper alloy is selected, galvanic effects can be avoided. Alternatively, the entire canister can be cold spray coated with CISCC-resistant materials.

2.3.6 Radiation Resistance

There is low risk of radiation damage to cold spray coatings due to the relatively low doses being emitted from DCSS canisters. Cold spray coatings should have similar radiation resistance to forged or extruded metal of similar chemistries. Some variation may occur because of differences in the microstructure of cold sprayed and forged or extruded material. Testing can be done to verify at what dose limit radiation effects become a concern for cold sprayed materials and how it compares to extruded and forged material of the same chemistry. However, radiation testing may not be unnecessary for DCSS applications because of low neutron flux through the canister.

2.4 Application

Cold spray is appropriate for in situ repair/mitigation of existing canisters, new canister fabrication, and final disposal storage systems. Cold spray can be used to deposit metal that acts as a corrosion barrier over fabrication welds and their HAZs or to coat the entire outer surface of canisters. Cold spray will have a relatively easy path to commercialization, because of the significantly reduced regulatory requirement of coatings compared to other processes, such as welding, for DCSS canisters.

2.4.1 Surface Preparation

Surface preparation is important to ensure high adhesion. Grit blast, wire brush/wheels, scotch bright, grinding, machining, and low-velocity spraying with cold spray powder are all methods that can be used for surface preparation. Cold spray can be done without any surface preparation, but adhesion values will be significantly reduced.

For factory cold spray, grit blasting is a common surface preparation method. Blast medium material and size need to be carefully selected to avoid embedding of blast medium in the substrate. For field cold spraying, Scotch-Brite™ or a wire brush/wheel work well. For robotic crawler repair application in rad environments, cold spray powder at low velocity, such that it acts like grit blasting media, is the simplest solution relative to robotic crawler design. However, this method generates powder that does not stick to the substrate and may need to be cleaned up. Integrating machining or grinding wheel systems into the robotic crawler is feasible and reduces the waste generation. If robotic crawlers are designed to grind out machine damaged areas, they can be used for surface preparation with minimal or no modification.

2.4.2 Methods of Application

For factory HPCS of new canisters, large industrial systems with liquid-cooled nozzles can produce high-quality cold spray deposits economically. Helium recycling in factory cold spray enables improved quality at reduced cost (Howe 2017). Similar processes can be used in a dedicated processing facility for spent nuclear fuel canisters.

Field equipment for HPCS is a relatively new development by Army Research Lab and VRC Metal Systems. Recently, Army Research Lab has demonstrated HPCS with clearances as small as 1.5” using blended Ni and CrC-NiCr powder with helium carrier gas. Porosity less than 0.5% and bond strength of 29 ksi was achieved (Nardi et al. 2019). The nozzle shown in Figure 5 was used. Nozzles designed for very low clearances have a bend that results in reduced velocity of the gas stream. The exact same material sprayed with a straight nozzle has 38 ksi adhesion compared to 26 ksi achieved using the angled nozzle.

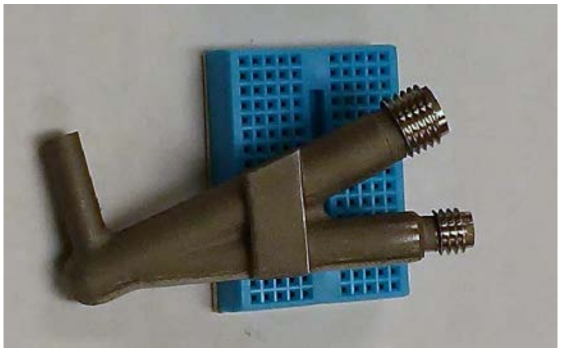

Figure 5. Cold spray nozzle capable of spraying a 1.5" inner diameter of a pipe (Nardi et al. 2019) designed by Army Research Lab. Image provided by Army Research Lab.

As part of a Small Business Innovation Research award from the DOE, VRC Metal Systems and their team demonstrated the ability to deposit SS and Inconel powders on SS 304L such that galvanic potential is matched and resistance to pitting is improved. A robotic crawler in a confined environment, representative of the space between a DCSS canister and overpack, executed HPCS using crude manual controls as a proof of concept demonstration. This work established the technical viability for cold spray mitigation and repair within the overpack using remote robotic equipment. Results are shown in Figure 6 below.

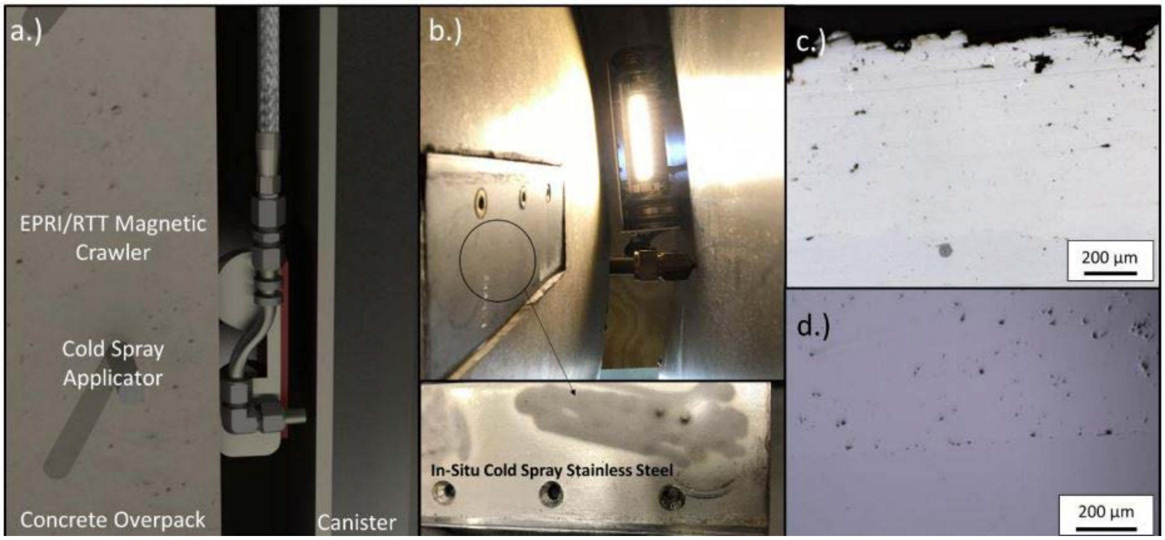

Figure 6. (a) Graphic of in situ cold spray repair, and (b) photo of cold spray mockup trail with Electric Power Research Institute - Robotic Technologies of Tennessee robotic crawler showing the viability of in situ stainless steel cold spray (c) cross section of cold sprayed SS 316L. (d) cross section of Inconel 625. Provided by VRC Metal Systems.

For situations where tight clearances do not exist, portable cold spraying can be done by manual handheld operation. Alternatively, the processes can be controlled using computer numerical control (CNC) by mounting portable equipment on a portable industrial robot arm.

2.5 Degradation Mechanisms

When best practices are followed, HPCS coatings can far outlast the austenitic SS base metal. This is because more resilient materials, such as nickel, can be used and compressive residual stresses are induced. The interface between the cold spray coating and base material that is exposed to the environment is an area of concern. Coating material needs to be properly selected and tested to ensure significant galvanic potentials do not exist between the coating and substrate materials.

Geometric effects need to be considered to ensure crevices or detrimental surface textures are not produced. The presence of crevices or rough surface textures could accelerate the onset of CISCC. All instances of CISCC in the fielded nuclear applications reported by the Pressurized Water Reactor Owners Group (Hosler and Hall 2010; Lareau 2014) were associated with crevice corrosion. This can be avoided by cold spraying to seal crevices.

Surface texture is affected by process parameters and powder preparation.

2.6 Cold Spray for life extension recap

HPCS is appropriate for in situ repair/mitigation of existing canister and new canister fabrication. HPCS can be used to deposit metal that induces compressive residual stresses and acts as a corrosion barrier over fabrication welds and their HAZs. Nozzles that are capable of spraying in areas with clearances as small as 1.5 in. are developed (Nardi et al. 2019) (Figure 5). Forces and temperatures required during the cold spray application are very low. Regulatory requirements for coatings are light compared to other technical areas, such as design or welding, for DCSS canisters. This will enable faster commercialization of the cold spray technology.

Benefits of cold spray include the following:

- Excellent mechanical properties can be achieved (>100 ksi [689 MPa] tensile strength, >30 ksi [207 MPa] adhesion strength).
- There is no deposition thickness limit.
- Below 1% porosity can be achieved. No interconnected porosity is produced.
- HPCS produces a high-density, high-hardness, and cold-worked microstructure with compressive residual stresses as opposed to the tensile residual stresses associated with melt/fusion based processes. This retards crack propagation and improves the CISCC resistance like shot peening.
- The process is inspectable using standard NDE techniques (Glass et al. 2018).
- Factory-based, field portable and robotic crawler cold spray systems are demonstrated or fully commercialized configurations.

Successful cold spray applications in other industries have resulted from significant research efforts and understanding to optimize the cold spray process for specific applications. Nevertheless, a significant amount of work needs to be done to develop HPCS for DCSS canister repair and mitigation, including the following research activities:

- Optimize coating chemistry for CISCC resistance and avoidance of galvanic effects
- Verify coating integrity in canister sliding and scraping conditions
- Develop an understanding of how surface roughness/texture developed by cold spray affect CISCC initiation.
- Reduce nozzle clogging and improve deposition efficiency for in-situ repair
- Optimize process for stationary equipment
- Optimize process for field repair conditions
- Quantify mechanical and thermal properties

- Additional mechanical testing to support structural modeling and analysis of cold spray coated components

The work above will result in cold spray process parameters and chemistries designed and optimized for CISCC repair and mitigation of DCSS canisters. Additionally, execution of the work above will enable: a broad understanding of the cold spray processes relative to CISCC resistance, identification of optimal material systems, documentation of best practices and technical datasets needed by stakeholders to implement, evaluate and regulate cold spray repair and mitigation of CISCC. A more thorough discussion of needed development work is included in sections 5-8.

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3 RELATED EFFORTS

This section describes related efforts relative to use of HPCS for canister life extension.

3.1 VRC Metal Systems Small Business Innovation Research Award

As part of a Small Business Innovation Research (SBIR) award from the DOE, VRC Metal Systems and their team demonstrated the ability to deposit SS and Inconel alloys on SS 304L such that galvanic potential is matched and resistance to pitting is improved. A robotic crawler in a confined environment, representative of the space between a DCSS canister and overpack, executed HPCS using crude manual controls as a proof of concept demonstration. This work established the technical viability for cold spray mitigation and repair within the overpack using remote robotic equipment. Results are shown in Figure 6 in section 2.4.2.

3.2 EPRI Residual Stress Measurements

The Electric Power Research Institute's (EPRI's) Extended Storage Collaboration Program (ESCP), identified cold spray to be a potential mitigation and repair technology of great interest (Tatman 2018). Because a primary factor that contributes to CISCC in susceptible materials is the presence of residual tensile stress, EPRI conducted a study of the residual stress conditions of cold sprayed Inconel 625 onto a weldment representative of a canister fabrication weld. The plates were mounted on a stiff strong back and then welded together. The strong back is sufficiently stiff to withstand the stresses created during arc welding. Inconel 625 was then cold sprayed over a portion of the weldment, as shown in Figure 7.

Residual stresses were evaluated and quantified using both contour and shallow hole drilling residual stress measurements. For comparison purposes, the as-welded (before cold spray) residual stress conditions were also evaluated. Hole drilling stress measurements of the as-welded mockups indicated longitudinal and transverse tensile residual stress of values of up to 621 MPa and 394 MPa. In contrast, in the cold spray on the highly strained weld mockup region, highly compressive stress values (as low as -1,100 MPa) were observed. Little change in the residual stress was seen in the as-welded region and within proximity to the cold spray buildup. Detailed results can be found in the EPRI report (Tatman 2020).

The results indicate that the stress reduction arising from cold spray could be as effective as common peening methods that are used for stress mitigation in the nuclear industry, further providing evidence that cold spray could be a beneficial repair method that assists future prevention of CISCC.

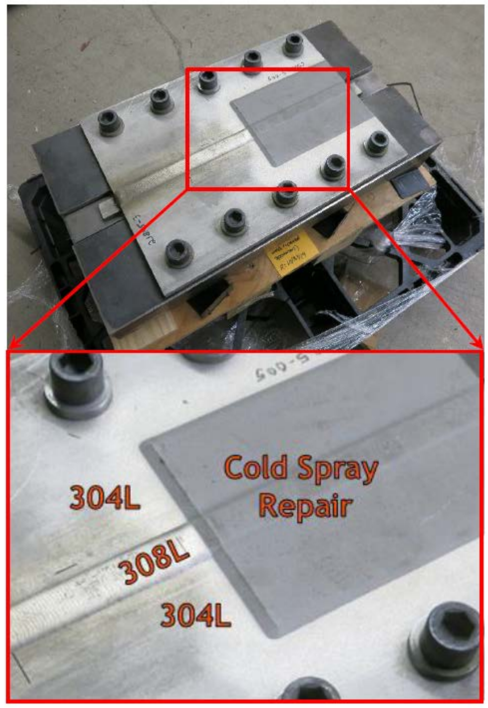

Figure 7. Inconel 625 cold spray buildup deposited on a high restraint weld mockup (Tatman 2020).

3.3 Nuclear Energy University Programs

Nuclear Energy University Program projects related to cold spray of canisters have been awarded to the University of Wisconsin Madison (UWM), Purdue University, and University of Cincinnati. Our project team is staying apprised of these efforts, providing technical guidance, and preventing duplication of efforts. Purdue is testing Inconel 625 sprayed with nitrogen and SS 304L sprayed with helium. University of Cincinnati is investigating laser-assisted cold spray. The UWM team is investigating cold spray of SS 304L and has published mid-project results (Yeom et al. 2020).

3.4 Hanford Cesium Strontium Dry Cask Storage

The Management of Cesium and Strontium Capsules (MCSC) project at the Hanford Site is building a dry storage facility on the Hanford Site to house cesium and strontium capsules (CSCs) in a DCSS that is a modified commercial spent nuclear fuel DCSS. Pursuant to achieving a 300-year design life for the canister, the project team has decided to coat fabrication welds and their heat affected zones with cold spray to create a corrosion barrier and induce compressive residual stresses to mitigate CISCC. Hanford contractors are working with the canister vendor NAC International, a cold spray vendor, and PNNL to develop the cold sprayed coating.

A lead canister will be fabricated, and heaters installed to mimic heat loading from a stored CSC which is representative of the nuclear waste. The lead canister will support aging management for MCSC and enable inspection of the canister without dose hazards. Portions of the weldments on the lead canister will be left uncoated for comparison purposes. In addition, test plates will be installed in the canister to enable testing and benchmarking of repair and mitigation processes within the overpack.

The lead canister and test plates are gaining interest across the industry. PNNL and EPRI are actively participating in the design of both. Stakeholders across the industry have expressed interest in fabricating various identical test plates using various processes (arc welds, various cold spray coatings, friction stir, and others) and locating them at multiple sites. Test plates located at various sites and under different conditions could provide valuable data to evaluate the long-term performance of various fabrication and mitigation techniques.

3.5 San Onofre Nuclear Generating Station

The San Onofre Nuclear Generating Station (SONGS) is a three-unit nuclear power plant located south of San Clemente, CA. The SONGS site currently has two Independent Spent Fuel Storage Installations (ISFSIs), with one horizontal storage system TN NUHOMS and one vertical storage Holtec UMAX system, that store the fuel assemblies decommissioned from this power plant. For the UMAX system, the Inspection and Maintenance require remediation measures if the storage unit does not ensure sound physical condition for on-site transfer or off-site transport.

Southern California Edison leveraged VRC Metal Systems SBIR work and expanded on it to demonstrate cold spray repair capability within a vertical canister overpack. This effort, leveraging expertise from participating parties, successfully developed the acceptance criteria for the cold spray process, and tested and implemented the robot/tooling package on a dimensionally correct mockup, enabling a remote robotic mitigation strategy for the canister storage. It was demonstrated that CP nickel with chrome carbide can be deposited using this development on the vertical canister (UMAX), if mitigation is required. Demonstration on a horizontal system is in progress at the time of the conclusion of this report.

3.6 Hanford Tank Farms

At DOE's Hanford Site, mild-steel waste tanks that have one-million-gallon storage capacities are used to store hazardous waste generated during the production of nuclear materials for defense applications. These waste tanks are buried below ground and have limited access ports. Many of these waste tanks are in service well beyond their original design life. Hanford tanks with design lives of 20 to 50 years have been in service for 55 to 70 years. The hazardous waste creates challenges for corrosion, and past corrosion control methods have not been fully successful. Pitting corrosion and wall thinning have been detected in the existing double-shell tanks (DSTs) used to store nuclear waste at Hanford (DOE 2019). Hanford has lost a million gallons of storage capacity due to leaks formed in the bottom of the 241-AY-102 DST primary tank (Johnson 2018). In addition, significant degradation has been detected in Hanford DST 241-AY-101 in the primary tank sidewall at the liquid-to-air interface (Johnson 2018).

Repair operations are made significantly more challenging than other industrial applications because of the hazardous radioactive environment and limited access. Leakage of waste can pose a significant risk to the environment, worker safety, and project schedules. Construction of replacement infrastructure will increase the footprint of contamination and divert funding and resources from the cleanup effort, thereby increasing the total cost and extending the schedule for the DOE Office of Environmental Management (EM) mission. The EM cleanup mission is currently estimated to extend beyond the next 60 years with completion estimated to occur from 2080 to 2200 (DOE 2019).

Cold spray is a solution of interest for repair of damaged tanks. A modest set of cold-spray conditions were tested to determine if a set of cold spray parameters exist that lead to dense, robust deposits of mild-steel powder on a mild-steel plate typical of what's found in Hanford tank farms (tanks and pipes). The tests included the deposition of cold sprayed carbon steel material to a pitted surface (simulated flaw) to demonstrate the ability to fill pits and restore wall thickness/corrosion allowance. The tests also included the deposition of carbon steel material to a flat surface opposite a pitted surface (test mockup) as an alternative means of restoring wall thickness/corrosion allowance.

Test results demonstrated mild-steel powder could be bonded to mild-steel substrates (i.e., similar powder/substrate material bonding) via the cold spray process using commercially available equipment operating within the range of normal process conditions. Measurement techniques used to characterize the cold-spray deposits demonstrated deposit densities greater than 99% (<1% porosity). The best results relative to deposit density, void distribution, and visual appearance were achieved using helium gas at an operating temperature of 500°C and a supply pressure of ~4.8 MPa (~700 psi). Determination of whether lower deposit densities could be used to create an acceptable repair is beyond the scope of this work. Characterization of the hermetic sealing and powder adhesion as a function of deposit density and thickness as well as the corrosion resistance are areas of study for future work.

The feasibility project satisfied the go/no-go criteria that were established to determine whether cold spray should be considered a candidate tank repair method and, therefore, whether future work would be warranted. Future efforts would (1) identify the range of cold-spray parameters and surface conditions that will lead to dense, robust deposits, and (2) adapt a commercial cold-spray system to perform tank/pipe repairs in an operational environment.

Commercially available mild-steel powder was annealed to soften the material such that bonding between the powder and the substrate could be achieved. In addition, screening to remove fines below ~20 µm was performed. Future work is recommended to evaluate modifications to the powder properties that may obtain bonding at reduced pressure or temperature to reduce helium usage. Final powder specifications can be provided to powder vendors to allow production runs of ready-to-use powder to be generated.

Surface preparation impacted deposit density and appeared to affect bonding/adhesion at the substrate interface and throughout the layer of deposited material. Limited testing demonstrated that the cold-spray process equipment could be used for surface preparation to enhance material bonding. Additional information is published in the WM2020 proceedings (Enderlin et al. 2020).

4 TECHNOLOGY DOWNSELECTIONS

Practices and principles for product development/design can be leveraged in technology selection and development for DCSS life extension techniques. While descriptions and implementation of best practices may vary, they can be broken down into the following steps:

1. Establish needs.
2. Develop understanding.
3. Generate concepts.
4. Score concepts.
5. Test concepts.

The following sections discuss two examples of technology down selections relevant to extending the life of DCSSs. A discussion of how principles of product development and design can be applied to achieve program objectives follows.

4.1 PNNL Technology Down Selection for Hanford Tank Repair

As described in Section 3.6, some DSTs used to store nuclear waste at Hanford Site have been in service beyond their intended design lives and exhibit significant degradation. An internally funded PNNL evaluation and down selection was done to identify repair processes most likely to succeed in restoring and extending the life of the Hanford tanks. A report about this effort is presented in Appendix A. The ensuing discussion demonstrates how the down selection process established needs, demonstrated understanding, generated the concept, and scored the concept. Concept testing is ongoing and initial results are discussed in Section 3.6 of this report.

High level process attributes identified in this down select of technologies include remote deployment feasibility, operational stability, deposit properties, substrate impacts, verification, technology accessibility and economics. Although technical details may vary, high level attributes identified and technology evaluations presented here are pertinent to repair and mitigation of DCSS canisters.

4.1.1 Establish Needs

In the absence of a set of functions and requirements for an EM mission infrastructure life extension system, PNNL staff formulated an initial description of the needs for a repair system to extend the service life of infrastructure components associated with the EM mission. The description of storage tank system needs was used as guidance for defining the problem to be resolved and the associated limits to aid the evaluation group in identifying related applications and potential technologies that could meet the need for a repair system.

A repair system is needed for building back material thickness at locations thinned by corrosion degradation and wear, sealing through-wall corrosion damage, and mitigating SCC problems identified broadly across the DOE complex. The repair of highest priority is considered the mitigation of material thinning occurring for in-service components that have yet to fail or the buildup of material to address potential wear associated with changes in processes. The objective of this repair is to build-back material in identified weakened locations to prevent component failure and extend the service life of components to meet EM mission needs.

The repair method needs to create a hermetic seal that can withstand the radioactive and corrosive environment for a relatively long period (>40 years). The repair methods sought are for sealing and corrosion/wear resistance and are not for repair of structural integrity. While a number of components may require repair in the future, the description of system needs emphasized the repair of underground waste tanks and transfer/process piping.

The evaluation group generated the following description of system needs:

- The repair technology needs to be applicable to mild-steels and SS, accommodate older metals no longer in production, allow repairs/material buildup for corroded surfaces, mitigate full material penetrations (e.g., leaks), and be applicable for repairing a wide range of materials. The desire is for the repair system to have the capability to mitigate an active leak under hydrostatic pressures of up to 60 feet of head (~35 psig for a 1.3 specific gravity fluid).
- The system should be capable of adding/building up a minimum of a half-inch of material. For this initial evaluation, no material buildup rate was considered. While solid material patches are a potential solution, they would need to be remotely transferred to the repair location. Therefore, additive processes (i.e., additional material is incorporated/added to the repair location as part of the process) are considered advantageous. The ability to apply dissimilar metals to improve corrosion resistance is desired.
- The repair system needs to operate remotely in a radioactive environment with umbilical lengths of up to 120 feet. The deployment will require insertion through cylindrical risers and must be capable of navigating multiple 90° turns. Access clearances of 12 in. are required.
- Beyond surface preparation (e.g., cleaning, adding a bonding agent), the repair method should require minimal material removal (e.g., drilling holes) or have minimal impact on the existing substrate (e.g., melting of substrate). Processes that require minimal or no surface preparation are preferable. If surface preparation is required, systems that can use the same equipment/repair process to conduct the required surface preparation are favorable. The need for deployment of a secondary technology to conduct the surface preparation is considered very disadvantageous.
- The repair needs to be verifiable and distinguishable from an unsatisfactory repair. Current assessments of tank integrity are performed via visual inspection and nondestructive examination using ultrasonic methods. Therefore, repairs that can be assessed and verified as being acceptable via these ultrasonic techniques are favorable.
- Because of the potential for flammable gas (e.g., hydrogen from radiolysis), applications with lower exposed temperatures (<470°C, ~80% of 585°C, which is recognized as the ignition temperature for hydrogen in air at atmospheric conditions) are desired, which do not require heating the substrate.

4.1.2 Develop Understanding

To identify and understand the potential candidate repair technologies, repair techniques applicable to flaws and leaks in metal structures and mitigation of corrosion damage were sought via literature review and discussion with vendors. In compiling a list of potential candidate technologies/methods, the description of need described above was not initially used to screen potential technologies. Therefore, techniques described as either temporary or permanent repairs were identified and put forth for consideration. Technical areas reviewed to obtain candidate technologies/methods included mitigation of CISCC in DSCC for spent nuclear fuel, techniques identified for commercial tank/vessel repair, and emergency repair methods for ship/boat leak repair as well as pipeline repairs. Learnings are summarized in Table A-2 in Appendix A.

The evaluation group identified 26 separate technologies/methods as potential repair methods. These technologies/methods are listed in the second column of Table A-2, with a brief description of each technology provided in the third column. The 26 technologies were grouped into four categories and 12 high-level processes, which are indicated in the first column of Table A-2. The four categories consist of Coatings, Solid Phase Processing (SPP), Fusion Processing, and Non-process Repairs. A review of Table

A-2 and the text that precedes it demonstrates deep understanding of relevant repair processes as they relate to project needs.

Understanding is evidenced by descriptions and categorization that simplify analysis and enhance understanding, as described in Table A-2. Table A- demonstrates deeper understanding of needs by organizing them into high-level attributes with associated factors and weights.

4.1.3 Concept Generation

Developed understanding was used to select proposed solution concepts based on deeper understanding of needs and technologies developed. A preliminary screening was conducted to determine if any of the high-level processes from Table A-2 should be eliminated from the head-to-head comparison. The following criteria were put forth as guidelines for the initial screening:

- Repairs are to be considered long term and semi-permanent (lasting 40 years).
  - Note: No repair was considered permanent because of the potential impacts of the corrosive and radioactive environment. To assign a designation of permanent repair would require further assessment.
- Repairs are not to rely on non-metallic materials that have not undergone dose testing to assess service life in the radioactive environment.

Based on these two criteria, the following processes were eliminated from further consideration: the category "Non-Process Repair," consisting of the high-level processes of mechanical and adhesive methods, and the high-level process "non-metallic powder coatings," from the "Coating" category. These three high-level processes were excluded because of the use of non-metallic materials in the repair, for which application to radioactive environments was not currently known and further investigation was considered beyond the scope of this initial down-select process.

In considering the high-level process "gas state coating," from the "Coating" category, the evaluation group deemed it impractical because of complexities associated with setting up and conducting the process for remote application, and therefore eliminated the process from further consideration.

Therefore, the preliminary screening reduced the number of high-level processes to be carried forward for a head-to-head comparison from the original 12 to the following 8:

- solution-state coating (cold spray)
- liquid-state coating (thermal spraying)
- solid-state coating (considered both a coating and a solid phase process)
- solid phase processing (friction stir welding)
- laser welding – fusion process
- powder processing – fusion process
- arc welding – fusion process
- electron beam welding – fusion process.

4.1.4 Concept Scoring

A subjective weighting was assigned to each attribute based on the anticipated priority of the attribute as applied to this application. The higher the percentage assigned to the attribute, the greater the importance (i.e., weighting) given to that attribute. The methodology applied used a percentage weighting such that a perfect score for a technology would equal 100.

The assigned weightings for each high-level attribute are listed in Table 2 and Table A-3 and are based on the subjective scale provided in Table A-3. "Operational suitability" and "properties of deposit" were identified by the evaluation group to be of highest priority and each was given a weighting of 20% of the final score, (i.e., "Very important" per Table A-3). While the "ability to deploy remotely" and "verification of the repair" were considered high-priority items, they were weighted less, 15%, because engineering development could potentially improve these attributes for this specific application. In comparison, the "operational suitability" and "properties of deposit attributes" are considered related to the physics and process methodology, regardless of the specific application. The "substrate impact" was considered similar in importance to the "ability to deploy remotely" and "verification of the repair" and was thus also assigned a weighting of 15% of the final score. Due to the nonproduction, non-commercialization, and limited application aspects of the repair process, "economics" was viewed as the least important attribute and was assigned a weighting of 5% of the total score. Because of the unique application of the repair technology and limited applications in the nuclear waste environment, the attribute of "technology accessibility" was given a weighting of 10%, which is less than all others except "economics."

The individual attribute factors were scored on a scale of 0 to 5. The subjective scale applied for scoring the individual attribute factors is also presented in Table A-3. For each attribute and candidate technology, the scores for the associated attribute factors were summed and a relative score was calculated based on the percent weighting for the individual attribute. However, if a score of 0 was given a single attribute factor, the associated attribute was given a score of zero for that technology. For example, the attribute "Substrate impacts" has five attribute factors listed in column three of Table A-3. If each attribute was given a score of 5, the sum of the attribute factors would be 25 (i.e., a perfect score), but the relative attribute score would be 15 (i.e., equal to the weighting percent of the attribute; refer to column 3 of Table A-3). Considering the same attribute, if each attribute factor was given a score of 3, the sum of the attribute factors would be 15, but the relative attribute score would be 9. However, if any single attribute factor was given a score of 0, then the relative attribute score would default to 0.

The scoring of the technologies was conducted independently by each member of the evaluation group. The attribute scores applied by each evaluation group member were whole numbers, 0, 1, 2, 3, 4, or 5 (i.e., no decimal values used). The average of the individual relative attribute scores was calculated from the individual relative attribute scores of evaluation group members for each technology and these scores are presented in Table 2. The total scores presented in Table 2 are the sum of the averaged relative attribute scores. Cold spray was ranked the most promising technology to pursue for EM mission infrastructure repair, with a relative total comparative score of 76.6.

Table 2. Results of comparative evaluation for candidate repair technologies.

Categories[a, b] =>		Coating		Solid Phase Process		Fusion Process			
High-level Processes[c] =>									
Attribute	Attribute Weighting (%)	Solution-state Coating	Liquid-state Coating (thermal sprays)	Solid-state Coating[d]	Friction Stir Welding/ Processing	Laser Deposition / Laser Beam Welding	Powder Coating / Powder Bed Processing	Arc Welding	Electron Beam Welding
Ability to deploy remotely	15	4.2	11.6	14.2	5.2	7.4	4.6	11.6	3.8
Operational stability	20	3.1	10.2	18.0	16.0	9.1	6.0	10.9	8.9
Deposit properties	20	7.2	7.2	11.7	12.3	8.3	6.9	7.2	6.7
Substrate impacts	15	10.6	8.0	12.8	12.6	7.2	7.6	5.8	6.8
Verification	15	9.0	9.0	10.0	12.3	8.7	8.0	6.7	8.7

Technology accessibility	10	7.3	6.9	6.7	4.9	6.4	4.9	10.0	5.3
Economics	5	2.2	3.5	3.2	2.5	2.7	2.8	4.7	2.5
Total Score:		***43.6***	***56.4***	***76.6***	***65.8***	***49.8***	***40.9***	***56.8***	***42.7***

(a) The categories used for grouping high-level processes are listed in Table A-2.
(b) Preliminary screening eliminated the "Non-Process Repair" category from the comparison evaluation.
(c) High-level processes remaining for comparative evaluation after preliminary screening. Refer to Table A-2
(d) Solid-state coating processes are considered both coating processes and a form of solid-state processing.

For the concept scoring process, the attribute factors could be expanded or modified to tailor the comparative evaluation to a more specific application or environment. The scoring of the attribute factors is subjective but provides a relative comparison of the technologies for each single attribute. The weighting of the attributes to obtain a final score is subjective and needs to be evaluated by the end users. Using the same attribute factor scores, a different ranking of the technologies may be obtained based on weighting/prioritizing the attributes differently. For example, Table 2 shows that the attribute "ability to deploy remotely" is weighted at 15 out of 100. A perfect score from the team (all three members rating all attribute factors a 5) would be 15. The relative attribute scores range from 14.2 to 3.8, indicating that the team views some high-level processes as easier to deploy remotely than others. The choice of weighting is important because the total scores range from 76.6 to 40.9 out of 100, so there were no perfect scores. The PNNL team agreed that the attribute weights used in the evaluation were appropriate for infrastructure repair for the EM mission, but recognizes that differences in priorities (changing the weight of each attribute) could change the order in the overall ranking of the high-level processes.

4.2 EPRI ESCP Repair Task Group Technology Summary Tables

The down selection described in the previous section involved a thorough evaluation. In some instances, decisions must be made where there is less time or fewer resources available for analysis. As part of EPRI's Extended Storage Collaboration Program (ESCP) a repair task group was formed to evaluated repair methods for DCSS canisters. The repair task group operates on a consensus basis and consists of industry subject matter experts whose efforts are supported by their employers. One objective of the repair test group is to identify and evaluate methods/ practices (tools) to repair environmental degradation of DCSS canisters. Pursuant this goal the task group held a series of meetings and conference calls to establish needs, develop understanding, generate concepts, and score concepts/methods for repair of canisters. The results are recorded in technology evaluation tables shown in Appendix B. Table B-1 through Table B-3 show that scoring differs for repair within an overpack, with overpack removed and considering fast confinement only. Learnings from this effort enabled down selection of technologies of interest (Tatman 2018) to a smaller set of technologies for further investigation.

These consensus-based efforts rely on the existing knowledge, expertise, and judgment of those who participate. The task group included owner/operators, cask vendors, engineers, and technology experts.

4.3 Discussion

Technology down selection can be consensus driven, analysis driven, or experiments driven. Process development typically involves multiple down selections that progress from consensus (brainstorming) to analytical (modeling and concept design) and end with data-driven down selection (benchmarking).

Consensus-based down selection can be described as brainstorming. The EPRI ESCP Repair Task group Technology Summary tables are documented outcomes of multiple task group meetings that were effectively brainstorming sessions. Consensus-based down selection is the first step, because typically investments cannot be made in analytical work until stakeholders agree on scope.

The Hanford tank farm work is an example of an analytical down selection. Analytical work was done to describe relevant physics, principles of operation, and technical knowledge to support weights and rankings.

Benchmarking is the ranking of concepts based on performance data. Concepts are tested using standards and performance data are collected. For each test, the performance must be numerically quantifiable. The test result is multiplied by a weighting factor (based on the importance of the associated need). For each concept, the weighted scores from all tests are combined into a total score, which is compared relative to existing methods and competing concepts.

In commercial product development the test metrics by which designs will be benchmarked is typically known prior to the initial brainstorming. The metrics by which canister repair and mitigation techniques should be measured are not clearly defined. Furthermore, the required metrics and weighting of performance metrics are highly dependent on what type of canister repair is being performed. When the repair task group was first formed, it attempted to include scoring for all conditions in one table. It soon became apparent that needs for each repair condition (with the overpack, overpack removed, and fast confinement) are different enough to require dedicated tables.

Test standards and metrics by which technologies will be evaluated will be governed by stakeholder needs. It is important for stakeholders to understand that the technical development for portable cold spray in confined spaces (within the overpack) is in opposition to technical development of cold spray with the overpack removed because of conflicting needs, weightings, and process configurations that are discussed further in the future work section. Road mapping activities done next year will ensure program needs and priorities are fully understood, and target performance metrics are defined.

5 PREPARATORY WORK

Prior to fabricating test coupons, preparatory work was done to ensure meaningful data will be generated.

5.1 Criteria for Comparative Analysis of CISCC Performance

High-fidelity CISCC analysis and testing is cost- and time-prohibitive for down selection of top-performing processes and parameters. Therefore, accelerated corrosion testing, such as ASTM G36, ASTM G61-86 (ASTM G61-86(2018)), is to be conducted to benchmark the corrosion resistance of each material. Learnings from this testing will generate understanding of how process parameters affect microstructure and CISCC resistance and will inform additional processes and material system optimization work at PNNL.

This section discusses criteria required for comparative analysis of CISCC performance. For comparative CISCC analysis of cold spray chemistries and developed microstructures, test methods must (1) normalize surface geometry and expose processes microstructure, (2) account for residual stresses, (3) be in a controlled and appropriate test environment, and (4) be numerically quantifiable and repeatable. Failure to meet these criteria could result in outcomes that cannot be used for comparative analysis of processes and materials.

Once cold spray chemistries are optimized, surface geometry effects can be investigated. Powder processing, surface preparation and process parameters affect cold spray surface geometry. Effects of these parameters on surface geometry and CISCC performance is an important undertaking. Additionally, it is important to quantify CISCC performance of buffed or ground cold spray materials. Test methods for comparing surface geometry effects must (1) account for residual stresses, (2) be in a controlled and appropriate test environment, and (3) be numerically quantifiable and repeatable. Failure to meet these criteria could result in outcomes that cannot be used for comparative analysis of processes and materials. These criteria can also be used for comparative benchmarking of materials and processes as they would exist in the field or process optimization after a material system has been selected. Without surface normalization, CISCC performance is a compound of chemistry, microstructure and surface finish. It may not be possible to decouple the effects of these factors unless prior testing was done with normalized surface geometries.

Learnings from the accelerated corrosion testing and processes optimization will be used to down select top-performing processes for detailed analysis and testing. Top-performing processes will be applied to coupons fabricated from a single heat of SS 304/304L. These coupons will be tested in four-point bending systems designed for CISCC testing at PNNL using direct current potential drop to perform in situ monitoring of crack formation and growth.

In addition, cold spray should be performed over arc welded test samples with and without laboratory generated CISCC cracks in the welds to simulate mitigating field repair conditions for CISCC. Arc-welded specimens without application of cold spray should also be tested as control samples. This will allow evaluation of the effectiveness of cold spray in mitigating both CISCC initiation and growth.

5.1.1 Normalization of Surface Geometry and Exposing Process Microstructure

Surface finish strongly affects CISCC and can sometime overshadow effects of chemistry or microstructure. Therefore, to understand chemistry effects on CISCC resistance and optimize a cold spray powder chemistry, surface conditions must be normalized.

For new fabrication, repair, or mitigation of canisters, welds are ground or machined flush. Many studies (Ghosh and Kain 2010; Turnbull et al. 2011; Acharyya et al. 2012; Lyon et al. 2015; Zhou et al. 2016;

Mankari and Acharyya 2017) have shown that machining or grinding of austenitic SS generally strongly affects CISCC resistance, as determined by ASTM G36. Several works (Turnbull et al. 2011; Lyon et al. 2015) noted that pitting and cracks *first formed along machining grooves and other surface disruptions*. This means that for the conditions reviewed in these studies, the surface condition appeared to be a leading factor for reduced CISCC resistance. Specifically, it is reported that very smoothly machined surface or ground surface featuring a thin work hardened layer with presence of micro-cracks and grain fragmentation can adversely affect CISCC resistance.

Section 1.2 of the ASTM G36 standard states "The boiling magnesium chloride test is applicable to wrought, cast, and welded SS and related alloys. It is a method for detecting the effects of composition, heat treatment, ***surface finish***, microstructure, and stress on the susceptibility of these materials to chloride stress corrosion cracking." The authors of the standard are stating that surface finish is a variable that affects the outcome of ASTM G36 testing.

Post processing, such as grinding or machining, cannot be used to normalize the surface. For a given end mill or grinding wheel, surface residual stresses, topology, and integrity are affected by processing parameters such as feed rate, depth of cut, and spindle speed. For a given machining or grinding operation applied at constant parameters, the surface topology, residual stresses, and integrity can still vary with material microstructures and/or other properties generated by the processes being evaluated. Therefore, grinding and machining cannot be used to normalize surface conditions.

Coupons should be electropolished such that the coupons meet a roughness tolerance and process microstructure is exposed. This allows processes to be compared without being obscured by surface geometry or post-process machining/grinding. It is important to note that mechanical polishing produces a nanocrystalline layer at the surface that is reported to be very corrosion-resistant and different from the original material (Wang et al. 2015). Therefore, processes such as electropolishing are preferred.

In one study (Mankari and Acharyya 2017) sanding was followed by buffing. This process produced a surface with reduced plastic strain, reduced surface roughness, and induced compressive residual stresses at the surface. ASTM G36 testing showed that this process significantly improved CISCC resistance relative to other conditions evaluated. These studies show that final surface processing can have a strong effect on CISCC initiation.

Arc welded and cold sprayed components are typically machined or ground after processing across all industries. For new fabrication, repair, or mitigation of canisters, it could be assumed that processed regions will be machined or ground. Based on the literature and understanding of relevant physics discussed in this section, it is reasonable to include a final material removal step, such as buffing, lapping, or polishing after the weld is ground or machined flush.

As far as the authors are aware, regarding cold spraying for nearly all applications, the top ~0.5 mm of the surface is removed. This is because it takes multiple layers of cold spray to fully densify material underneath. Even for cold spraying, it is important to normalize the surface by polishing to remove the transitory microstructure that exists in the top ~200 µm. However, for repairs within an overpack, removal of this top layer may be impractical and unnecessary. If needed, addition of a material removal process to stationary cold spray systems or robotic crawlers is doable.

Initial work will use machining followed by electropolishing for initial CISCC analysis so samples can be evaluated without confounding effects from surface geometry or material degradation (such as grain fragmentation or micro-cracks) produced by material removal processes.

Once top-performing processes are selected, various surface conditions can then be evaluated and optimized with and without post processing such as grinding or buffing.

5.1.2 Residual Stress

In order to successfully assess the mitigation efficacy of cold spray on CISCC, specimens must correctly represent the residual stress induced during different processes such as in welding (tensile residual stress) and after cold spraying (compressive residual stress). For example, U-bend samples that are bent after the application of cold spray cannot be used to correctly evaluate the effect of cold spray because the post-cold spray bending will induce tensile residual stress that compromises the beneficial effect of compressive residual stress induced by cold spraying. In addition, stress relief can occur when a weldment is cut, of which the magnitude varies with cutting location and intrinsic properties of the weld. Therefore, it is very important to make sure that the welded specimens retain similar residual stresses for eligible comparison.

5.1.3 Environment

Environment variables, such as salt concentration, humidity, and temperature, have a strong effect on CISCC and must be well-controlled during test and relevant to conditions of interest. Initial work will use ASTM G-36 defined procedures for chemistry and thermal conditions.

5.1.4 Result Quantification and Repeatability

Many CISCC test methods were developed for qualitative analysis. In order to effectively assess the impact of cold spray, methods need to be developed such that CISCC performance, specifically time to crack initiation, is quantifiable and repeatable as proven using gauge capability studies. High-reliability four-point bend testing in autoclaves is being developed at PNNL/SNL. This method should prove via gauge capability studies that results are sufficiently accurate and repeatable.

It is desirable to build upon ASTM G36 or other CISCC screening methods to produce a rapid screening study where results are numerically quantifiable and sufficiently repeatable. This should be proved with a gauge capability study.

5.2 ASTM G36 with Additional Controls

ASTM G36 is an accelerated test method of ranking the CISCC susceptibility for SS and related alloys. The materials being tested are immersed in a boiling magnesium chloride solution and are monitored periodically for examination of crack initiation. The aggressive nature of the boiling magnesium chloride expedites CISCC in materials, making it an effective method to quickly screen among a variety of materials. This test method is ideal for identifying the effects of composition, heat treatment, surface finish, microstructure, and stress on the CISCC susceptibility of materials.

ASTM G36 allows for a great deal of flexibility relative to how components are tested. To ensure repeatability and the criteria for comparative analysis, established in the previous section, are met the following additional procedures are added:

1. Coupons will be placed into four-point bend fixtures such that tensile residual stresses, representative of those produced by fabrication welds, exist on the exposed surface of the coupon.
2. Cold spray coating will be generated using varying processing parameters and deposited onto the convex face of the coupons after they are placed into bending.
3. Prior to testing, the side of the coupons that have been cold sprayed will be electropolished to normalize surface condition.
4. Accelerated screening test following the ASTM G36 will be performed on all coupons.

5. Coupons will be removed and inspected periodically. Time to first observed crack and total crack length at time intervals will be recorded. This will provide quantifiable test results that can be directly used to guide parametric optimization.

5.3 Large Plate CISCC Crack Fabrication for Repair Demonstration

PNNL previously developed and implemented a CISCC generation method for large plates. This is done by placing the plate into a four-point bending fixture and exposing a sealed section at the center of the plate to a boiling 6M $MgCl_2$ solution at 108°C. This novel method breaks the coupon size limitation of ASTM G36 and the time intensiveness of ASTM B117. The boiling magnesium chloride is temperature controlled by heaters on the back side of the crack generation specimen, and oxygen supply is maintained and controlled via pipe venting on the fixture; 108°C is low enough to avoid heat affected microstructure change during crack generation. This method allows for the development of CISCC cracks and corrosion products, enabling investigations of real CISCC crack repair via cold spray.

The fixture design is shown in Figure 8 below. The flexure fixture is a PNNL-built setup consisting of a 4” by 21” I-beam as the base of the setup. The plates to be exposed are then flexed over two 3” mandrels spaced 5” apart. The mandrels have two flat sides, reduced 0.25” in height, to accommodate mounting onto the I-beam, as well as, allowing the 2.5” wide heating fixture to be installed in the concavity of the plate flexure. The crack generation specimen is bolted at all four corners located 1” away from the edges. The fixture has a 4” wide steel plate bolted to the sides for support and as an anchor point for the lab jack. The lab jack supports the corrosion-resistant chlorinated polyvinyl chloride (CPVC) cup that holds the 6M magnesium chloride solution.

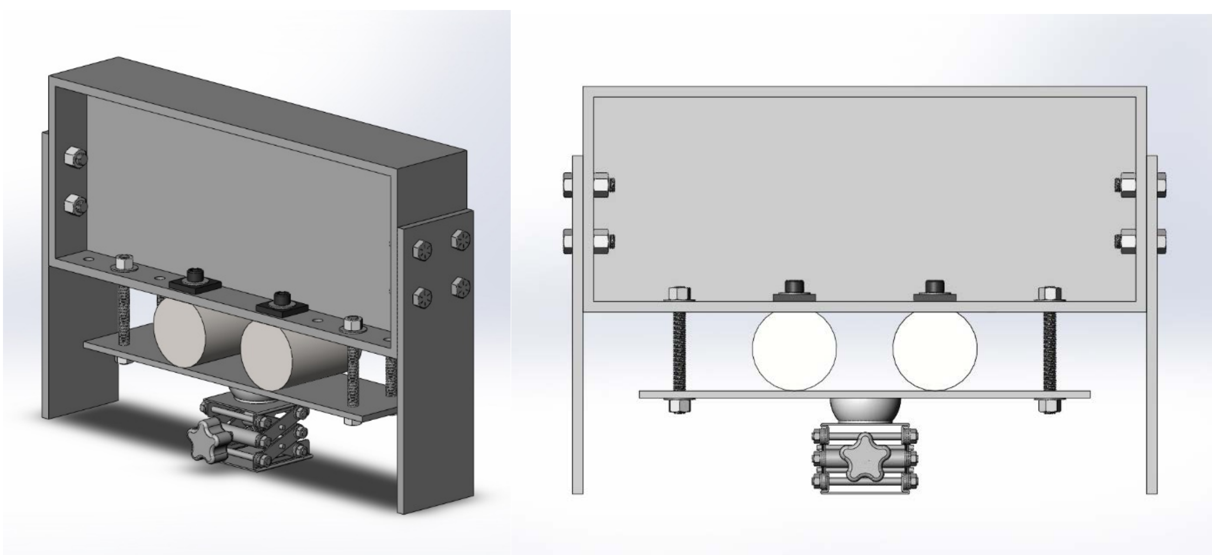

Figure 8. PNNL Crack Generation Fixture Set Up

The sensitized SS plates used for the CISCC crack generation were heat treated in a Thermal Technology Vacuum Furnace Model # 121224M MS. The sensitization treatment calls for a 600 °C soak for 40 hours in vacuum to minimize oxidation. The furnace was preheated to the target temperature and cooled down to room temperature both over a period of 120 minutes.

Rapid (24–48 hour) CISCC crack generation on sensitized SS plates is achieved by placing the plates surface under tension in a bend or flexure set up, exposing the apex to $MgCl_2$ solution at ~108°C; as shown in Figure 9 below.

Figure 9. Specimen in chloride crack generation fixture showing the full setup.

The apex of the plate is protected using VHT high temperature corrosion resistant paint (VHT Gray Primer SP148) except a long rectangular area in the center of the circle that is exposed to the $MgCl_2$ solution (as highlighted in silver in Figure 10). This exposed area is where the crack nucleation and propagation will occur.

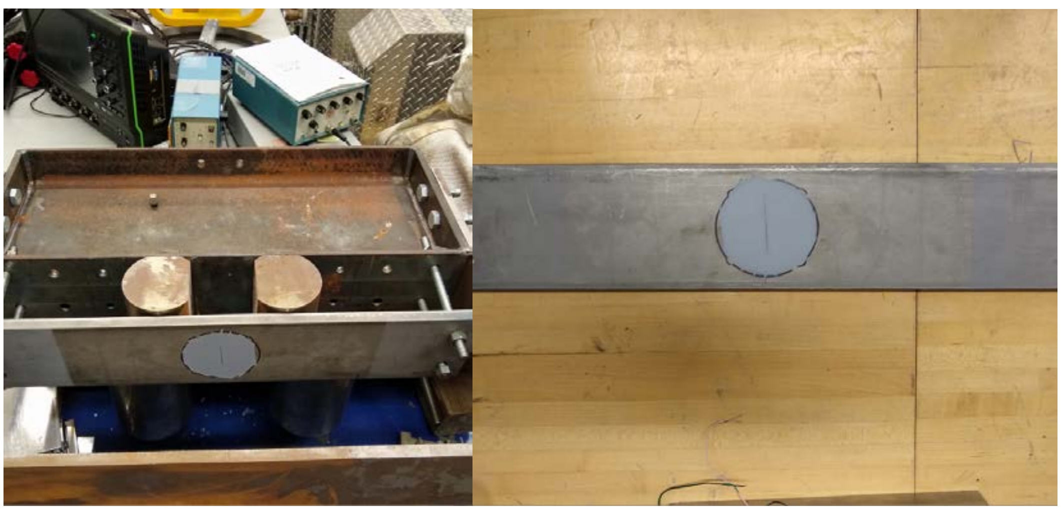

Figure 10. Specimen after 24 hours.

With a CPVC container clamped to the apex of the plate, 6M $MgCl_2$ is funneled into the container until all air is bled out. Once this is done, the plate apex is heated on the concave side using two-barrel heaters and a heating block. The heaters are controlled to the boiling temperature of the $MgCl_2$ solution and throttled to keep the $MgCl_2$ at ~108 °C for the duration of the exposure. The exposure starts when the $MgCl_2$ reaches the desired temperature. Notes are kept on test duration, temperature, and observations that may occur. Once the exposure is complete, a portable USB microscope is used to verify the presence and growth of cracks and to capture images. Figure 11 shows cracking in the sensitized plate within the sliver of unmasked area. After testing, the plate is removed from the flexure fixture and cleaned with solvent to remove the masking.

Figure 11. Micrograph of crack cross-section perpendicular to crack.

This method was used to demonstrate CISCC crack generation in an SS 304L plate. Additional CISCC bearing plates are being made to support cold spray repair work described in Section 6.2.3.

5.4 Powder Classification and Processing

Powder processing is important to producing high-quality cold spray coatings. All cold spray powders for the FY20 work scope have been received and processed. Powder processing includes:

1. Classifying powders to quantify particle size distribution.
2. Sieving powders to remove large and fine particles.
3. Reclassifying sieved powders to quantify particle distribution.

All powders ordered were "pre-sieved" powders. Powder classification showed that factory pre-sieving successfully removed fine particles (below ~5 microns). Additional sieving was done at PNNL to remove large particles. Table 3 below shows the standard deviation in powder size.

Table 3. Comparison of average particle size and standard deviations pre- and post- sieving.

		Super C	SS 316 L	Inconel 625	CrC 410
As Received	Average (µm)	39.32	33.27	33.84	28.91
	Std Dev (µm)	7.24	10.19	8.11	8.33
Dev Post-Sieving	Average (µm)	37.03	27.78	31.03	26.96
	Std Dev (µm)	5.67	5.74	4.98	6.05

6 FUTURE WORK

Due to delays and lab access limitations resulting from COVID-19, some work scope from FY20 will extend into FY21.

6.1 Completion of FY20 Scope

Cold sprayed test coupons will be made by depositing powders using materials listed in Table 4 on SS plate substrate using various processing conditions. Processing conditions refer to the combination of powder material selection, powder processing methods, equipment configuration, selected process parameters, carrier gas, nozzle design and other aspects that define how the cold spray was performed. Testing described in Table 5 will be performed for each powder material listed in Table 4 and mixtures of each alloy and 410 chrome carbide. Iterative coupon generation and testing will continue until the project team identifies optimal process parameters for high-fidelity DCPD CISCC testing and analysis.

Table 4. Proposed exploratory test plan for cold spray.

Grade	Alloy 625	410 Chrome Carbide	316LSS	Ni-Cr-Mo Alloy
C	<0.1%	~4%	<0.03%	0.10%
Cr	20–23%	Balance	16–18%	23%
Ni	Balance	~8%	10–14%	Balance
Mo	8–10%	-	2-3%	18%
Mn	<0.5%	-	<2%	-
Si	<0.5%	-	<0.75%	<1%
P	<0.015%	-	<0.045%	-
S	<0.015%	-	<0.03%	-
Cu	-	-	-	-
Fe	<5%	-	Balance	<1%
Al	<0.4%	-	-	-
Ti	<0.4%	-	-	-
Co	<1%	-	-	-
Nb	3.15–4.15%	-	-	-

Table 5. Testing and analysis to be performed on cold spray samples.

Criteria	Performer	Standard	Comments
Adhesion: Glue based Triple lug shear Tensile Specimen	PNNL	ASTM B571 MIL-J-24445A ASTM E-8	Glue based adhesion testing will be done for each spray condition. Triple lug shear and E-8 may be done for top performers.
Porosity	PNNL	Mil STD 3021	3 images per spray condition
Hardness	PNNL	ASTM E92	ASTM E384 (microindentation) may be used for blended powder coatings with hard particles.
Electrochemical	SNL	ASTM G61-86	3 replicates per spray conditions
CISCC resistance	SNL	ASTM G-36	Cold spray and tested while in four-point bend condition
Pitting	SNL	Coupons from each spray condition will be provided to SNL for including in ongoing pitting studies.	

Microstructure	PNNL	PNNL will execute microstructural evolution of each spray condition.

The effects of processing conditions and material selection will be understood from initial testing. High-fidelity DCPD CISCC testing and analysis will be performed on specimens with favorable conditions.

6.2 FY 21 Scope

This section describes work to be accomplished as FY21 work scope.

6.2.1 Process Optimization and Testing Support

Based on learnings from the FY20 work scope, cold spray conditions, material systems, and process parameters will be optimized through iterative experimentation and analysis. Additional test coupons will be made to support ongoing pitting and CISCC testing at SNL and PNNL.

6.2.2 Cyclic Thermal and Pressure Loading

In FY21, cold spray will be used to repair cracks in pipes and testing will be done to establish guidelines for the thickness of cold spray repair based on (1) properties of the cold-sprayed material produced by the selected material and parameters, (2) internal pressure, and (3) operating temperature. This work will be performed for conditions related to CISCC canisters in collaboration with other Spent Fuel and Waste Disposition (SFWD) tasks.

There is an extremely high likelihood of success based on work done by PNNL collaborator VRC Metal Systems. Figure 12 provides screen shots from a video provided by VRC Metal Systems of a cold spray repair of an active water leak at 1,000 psi. It is important to note that to effectuate this repair, material was built up around the active leak. For CISCC repair, there will be no active pressure and a flat repair can be made that easily holds the pressure.

Figure 12. Repair of active leak using cold spray by VRC Metal Systems. Left: water leak; right: final repair. Pressure of the pipe was 1,000 PSI during and after the repair.

6.2.3 Valid Properties of Cold Spray on Arc Welds and Cracked Arc Welds

Repair and mitigation demonstrations and process validation studies will be done to establish viability and present performance metrics for cold spray over arc welded plates and CISCC cracked plates with and without arc welds.

Cracked plates with and without arc welds will be generated using methods described in Section 1. Small four-point bend coupons with be cut with the generated crack centered on the coupon length running perpendicular to the length. Coupons will be placed in four-point bend fixtures designed for boiling magnesium chloride testing and loaded in tension. Coupons will then be cold sprayed and tested in accordance with ASTM G36. Additional coupons will be generated to perform all other tests described in Table 5 to establish the properties of cold spray on arc-welded material with and without CISCC cracks.

It is anticipated that performance of cold spray will improve when applied over arc welds because of the closer match in hardness/ductility of arc-welded filler metal and atomized cold spray powders.

6.2.4 Sliding and Scraping of Cold Spray against Concrete Overpack or Metal Rails

During transportation and installation, dry storage canisters experience wear from sliding along metal rails and scraping against the concrete overpack. It will be beneficial to evaluate the wear resistance of the canister's outer surface against the metal and concrete materials. Identifying suitable testing standards is needed to develop cold spray coatings that are not compromised by scraping or wear during canister installation. Numerous testing standards are related to wear, based on the wear mechanism, materials species, specimen shapes, and sizes.

Choosing suitable testing standards begins with identifying the wear mechanisms. In general, there are two types of wear, adhesive and abrasion, both of which could be affected by corrosion.

Adhesion wear occurs when two solid surfaces are in sliding or in rolling contact with each other. The sliding movements could result in atomic contact-asperities, and shear deformation between the contacts. Eventually, shear failure could occur.

Abrasive wear includes sliding abrasion and impact abrasion. Sliding abrasion takes place when particles or sharp irregularities on the surface scratch a softer surface and cause a volume loss or plastic deformation of the specimen. Two-body and three-body abrasion wear are two different forms of abrasion wear, distinguished by presence of wear-debris. Different scratching mechanisms can result from abrasion wear, such as micro-ploughing, micro-cracking, micro-fatigue, or micro-cutting. Often a combination of different scratching mechanisms exists in real applications. Impact abrasion/particle erosion occurs when a surface is being hit by solid particles, causing plastic deformation or materials removal.

Regarding the scenarios identified in the beginning of this section, the dry storage canisters typically experience adhesion wear and sliding abrasion. Corrosion does not occur during sliding or scraping but performing wear tests on laboratory corroded samples could provide an understanding of impacts of corrosion on canister or coating integrity. This is applicable to the removal of canisters.

Table 6 lists the relevant standards, the dominant wear mechanisms, and the application space of the standards.

Table 6. Possible test standards for evaluating sliding and scraping.

Testing Standards	Wear Mechanism	Application Space
ASTM G99 - 05 Standard Test Method for Wear Testing with a Pin-on-Disk Apparatus	Adhesion	Metal-on-metal
ASTM G132-96 Standard Test Method for Pin Abrasion Testing	Sliding abrasion	Possible for metal-to-concrete
ASTM G174-04 Standard Test Method for Measuring Abrasion Resistance of Materials by Abrasive Loop Contact	Sliding abrasion	Possible for metal-to-concrete

6.2.5 Cold Spray Technology Development Road map

Technical road mapping activities will begin with establishing priorities and associated target specifications. Defining the relative importance of types of cold spray applications is important because they have competing system configurations and objectives.

Cold spray done within an overpack is challenging due to space confinements. For cold spray in confined spaces, portable cold spray systems, which have remote gas heating, and angled nozzles are used. Remote gas heating limits spray gas temperature. Velocity reduction and associated angled nozzles require expensive helium gas to be used for all materials of interest for canister life extension. Velocity reduction and flow direction change dramatically reduce deposition efficiency to below 50% for materials that can be sprayed with stationary equipment at greater than 99.9% efficiency. Angled nozzles and lack of nozzles cooling cause nozzle clogging to occur quickly in portable systems. These issues are specific to cold spray in confined spaces that only apply to repairs within the overpack. Overcoming issues associated with angled nozzle spraying in a confined space is an important undertaking for development of repair within the overpack.

For all other cold spray operations, factory type equipment using straight nozzles and improved heating can produce high-quality coatings at a fraction of the cost per unit weight of deposited materials if developed correctly. Nozzle cooling, higher gas temperatures, and helium recycling provide process advantages and improved economics. Developing processes that leverage these advantages can enable favorable economics for coating entire canisters, or large areas of canisters after as part of DOE acceptance of canisters. Costs reduction is also of interest for application of cold spray over arc welds and heat-affected zones as part of new canister fabrication.

Road mapping activities will ensure that cold spray development activities optimize value to stakeholders and reflect Spent Fuel and Waste Disposition priorities despite the divergence of technical challenges between cold spray applications. Material system optimization, surface preparation, and other technical challenges being addressed in the current work scope apply to all cold spray applications for canister life extension.

7 OVER THE HORIZON COLD SPRAY TECHNOLOGY ADVANCEMENT

Structural and mechanical requirements need to be evaluated for thorough evaluation of the cold spray technology for this application. For example, does the coating need to survive being dragged across rails or scraping against concrete while being lowered into the overpack? If so, what are the laboratory test metrics that prove the coating can meet these needs? It is important to understand that coatings and base metals react loads together, not independently. Structural analysis, using FEA, and mechanical testing will likely be needed to understand the combined behavior of the coating and canister base metal. This understanding can inform coating property requirements.

Nozzle clogging is an issue in portable HPCS systems when spraying nickel and nickel-based alloys. Clogging can be solved by developing cooled nozzles for portable system or adding hard particles, such as carbides, to the powder. Hard particles can improve the mechanical properties of the deposited material but could cause localized galvanic effects that accelerate pit formation. Nozzle cooling and the effects of hard particles are areas that need to be investigated for applications using portable equipment.

A significant amount of work needs to be done to develop HPCS for DCSS canister repair and mitigation. Coating powder chemistries needs to be selected such that no detrimental galvanic effects occur and CISCC resistance is maximized. Identifying the optimal chemistry for canister protection is an area that demands a significant R&D effort.

Surface roughness/texture effects are expected to affect CISCC initiation. Cold spray parameters and powder preparation can affect surface roughness. Testing should be done to develop an understanding of how surface roughness/texture developed by cold spray affect CISCC initiation.

Edge effects and interfaces between the coatings and substrates exposed to the environment need to be investigated. For example, deposited coatings could produce geometric discontinuities that enable crevice corrosion. If this is the case, a groove and blend technique (Figure 13 bottom) can be used to normalize surface geometry. Alternatively, grinding or buffing edges of a deposited costing without a groove could be a solution.

Figure 13. Coating strategies for localized coatings. Top: deposited coating without groove or blend. Bottom: grooved and blended coating.

Process optimization to develop optimal nozzle design, surface preparation, powder preparation, and quality control/assurance techniques are needed after coating chemistry is finalized and geometric effects are understood.

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Appendix A

Repair Method Down Select for Repair of Hanford Storage Tanks

The following is the contents of letter (PNNL-SA-150668) sent to certain Hanford nuclear waste storage and processing stakeholders on January 24, 2020 describing a PNNL internally funded Technology down selection for repair of Hanford waste storage tanks. Appendix table numbers have been applied so the tables can be referenced properly in the body of the report.

INITIAL DOWN SELECT OF REPAIR METHOD FOR EM INFRASTRUCTURE LIFE EXTENSION CONDUCTED BY PNNL

This letter summarizes a down-select process conducted by an evaluation group composed of three Pacific Northwest National Laboratory (PNNL) staff members (Carl W. Enderlin, Christopher B. Smith, Ken A. Ross) to identify a potential method(s) to pursue for repairing and extending the service life of infrastructure within the U.S. Department of Energy (DOE) complex that is needed for nuclear waste storage and processing (e.g., waste tanks, transfer pipelines, evaporators) for the Office of Environmental Management (EM) mission. Results are presented in Table A-1.

To identify potential candidate options for conducting a technology down-select process, a common industry practice is for end users to generate a set of functions and requirements for the field deployment from which technology attributes can be identified for developing comparisons between various technologies and methods. These attributes can then be ranked or weighted to give priority to specific attributes based on the application's needs and requirements.

In the absence of a set of functions and requirements for an EM mission infrastructure life extension system, PNNL staff formulated an initial description of needs for a repair system to extend the service life of infrastructure components associated with the EM mission. The description of system needs was used as guidance for defining the problem to be resolved and associated limits to aid the evaluation group in identifying related applications and potential technologies that could meet the need for a repair system. Washington River Protection Solutions, LLC, incorporated aspects of the description of system needs into an industry expression of interest posted in late 2018.[1]

The PNNL evaluation group generated a list of candidate repair technologies and grouped these technologies into categories and high-level processes (Table A-2). The individual technologies/methods identified served as examples associated with each high-level process and the down-select process was conducted for the resulting high-level processes. A preliminary screening was initially conducted to determine which high-level processes should be considered for a comparative evaluation. Based on experience and the PNNL-formulated description of system needs, the evaluation group developed a set of attributes and specific attribute factors that could be individually scored for each high-level process to produce quantitative results for the comparative evaluation. The evaluation group assigned a percent weighting to each attribute based on a subjective determination of relative importance. The specific factors associated with each attribute were independently scored by each member of the evaluation group on a scale of 0 to 5 for each of the high-level processes included in the comparative evaluation. Table A-3

[1] Washington River Protection Solutions, December 2018. *Expression of Interest (EOI) for In situ Tank Repair for Infrastructure Life Extension.*

lists the attribute factors that were independently scored. Table A-3 provides the subjective score meanings used for scoring the attribute factors.)

The relative weighting of the individual attributes was formulated as percentages such that their sum was 100 and a perfect score for a process would equal 100. (Table A-3 lists the weighting factors that were applied to each high-level attribute.) For a given set of attribute factor scores, different results could potentially be obtained with a change in the percent weighting for the set of attributes.

This letter provides the results of the comparative evaluation conducted by the PNNL evaluation group, the PNNL-formulated description of system needs used to define the problem being addressed, a listing of the candidate repair technologies identified, and the grouping of the technologies into associated categories and high-level processes. In addition, the preliminary screening of high-level processes, the selected attributes, and comparative evaluation process are summarized.

Conclusions/Results

Based on the comparative evaluation and the formulated description of system needs, the evaluation group identified solid-state coating, which is considered both a coating and a solid phase process, as the most promising technology to pursue for EM mission infrastructure repair, with a relative total comparative score of 76.6. Solid-state coating consists of cold spray (supersonic nozzle velocities) and associated subsonic-related processes. While active leak mitigation was not an attribute factor scored for the comparative evaluation, it is a desired capability for a repair system per the evaluation group's formulated description of system needs. VRC Metal Systems LLC has applied cold spray to demonstrate the mitigation of a small, active leak in a stainless-steel pipe pressurized to 60 psig.[2] Table A-1 lists the categories and associated high-level processes identified for the comparative evaluation based on the preliminary screening, the attributes scored, the weighting assigned to each attribute, the comparative scores for each attribute, and the overall total scores for the high-level processes compared.

Friction stir welding/processing (FSW/P) was the second leading candidate based on the scored evaluation and associated attribute weighting, with a relative total comparative score of 65.8. While FSW/P is another solid phase process that compares well with the solid-state coating processes with respect to repair-related attributes, and development is currently under way to apply FSW/P to dry storage of spent nuclear fuel and waste packaging, deployment of FSW/P in the remote environments under consideration is more problematic due to the contact forces that must be applied by the friction stir tool to the substrate. The relatively high finish of FSW/P compared to other processes indicates that the attributes, attribute factors, and weighting of the attribute scores may need to be refined to specifically target in situ tank repair compared to other infrastructure repair.

[2] Video can be viewed on YouTube: *VRC Cold Spray Active Pipe Leak Repair*, https://www.youtube.com/watch?v=QsR7MnFCM74

Table A-1. Results of comparative evaluation for candidate repair technologies.

Categories [a, b] =>		Coating		Solid Phase Process		Fusion Process			
High-level processes [c] =>									
Attribute	**Attribute weighting (%)**	**Solution-state coating**	**Liquid-state coating (thermal sprays)**	**Solid-state coating [d]**	**Friction stir welding/ processing**	**Laser deposition / laser beam welding**	**Powder coating / powder bed processing**	**Arc welding**	**Electron beam welding**
Ability to deploy remotely	15	4.2	11.6	14.2	5.2	7.4	4.6	11.6	3.8
Operational stability	20	3.1	10.2	18.0	16.0	9.1	6.0	10.9	8.9
Deposit properties	20	7.2	7.2	11.7	12.3	8.3	6.9	7.2	6.7
Substrate impacts	15	10.6	8.0	12.8	12.6	7.2	7.6	5.8	6.8
Verification	15	9.0	9.0	10.0	12.3	8.7	8.0	6.7	8.7
Technology accessibility	10	7.3	6.9	6.7	4.9	6.4	4.9	10.0	5.3
Economics	5	2.2	3.5	3.2	2.5	2.7	2.8	4.7	2.5
Total Score:		***43.6***	***56.4***	***76.6***	***65.8***	***49.8***	***40.9***	***56.8***	***42.7***

[a] The categories used for grouping high-level processes are listed in Table A-2.
[b] Preliminary screening eliminated the "Non-Process Repair" category from the comparison evaluation.
[c] High-level processes remaining for comparative evaluation after preliminary screening. Refer to Table A-2
[d] Solid state coating processes are considered both coating processes and a form of solid-state processing.

Description of System Needs

A repair system is needed for building-back material thickness at locations thinned by corrosion degradation and wear, sealing of through-wall corrosion damage, and mitigation of stress corrosion cracking problems identified broadly across the DOE complex. The repair of highest priority is considered the mitigation of material thinning occurring for in-service components that have yet to fail or the buildup of material to address potential wear associated with changes in processes. The objective of this repair is to build-back material in identified weakened locations to prevent component failure and extend the service life of components to meet EM mission needs.

The repair method needs to create a hermetic seal that can withstand the radioactive and corrosive environment for a relatively long period (> 40 years). The repair methods sought are for sealing and corrosion/wear resistance and are not for repair of structural integrity. While there are a number of components that may require repair in the future, the description of system needs emphasized the repair of underground waste tanks and transfer/process piping.

The evaluation group generated the following description of system needs:

- The repair technology needs to be applicable to mild-carbon steels and stainless steel, accommodate older metals no longer in production, allow repairs/material buildup for corroded surfaces, mitigate full material penetrations (e.g., leaks), and be applicable for repairing a wide range of materials. The desire is for the repair system to have the capability to mitigate an active leak under hydrostatic pressures of up to 60 feet of head (~35 psig for a 1.3 specific gravity fluid).
- The system should be capable of adding / building up a minimum of a half-inch of material. For this initial evaluation, no material buildup rate was considered. While solid material patches are a potential solution, they would need to be remotely transferred to the repair location. Therefore, additive processes (i.e., additional material is incorporated/added to the repair location as part of

the process) are considered advantageous. The ability to apply dissimilar metals to improve corrosion resistance is desired.

- The repair system needs to operate remotely in a radioactive environment with umbilical lengths of up to 120 feet. The deployment will require insertion through cylindrical risers and must be capable of navigating multiple 90° turns. Access clearances of 12 inches are required.
- Beyond surface preparation (e.g., cleaning, adding a bonding agent), the repair method should require minimal material removal (e.g., drilling holes) or have minimal impact on the existing substrate (e.g., melting of substrate). Processes that require minimal or no surface preparation are preferable. If surface preparation is required, systems that can use the same equipment / repair process to conduct the required surface preparation are favorable. The need for deployment of a secondary technology to conduct the surface preparation is considered very disadvantageous.
- The repair needs to be verifiable and distinguishable from an unsatisfactory repair. Current assessments of tank integrity are performed via visual inspection and non-destructive examination using ultrasonic methods. Therefore, repairs that can be assessed and verified as acceptable via these ultrasonic techniques are favorable.
- Due to the potential for flammable gas (e.g., hydrogen from radiolysis), applications with lower exposed temperatures (< 470°C, ~80% of 585°C, which is recognized as ignition temperature for hydrogen in air at atmospheric conditions) are desired, which do not require heating the substrate.

Candidate Repair Technologies

For identifying potential candidate repair technologies, repair techniques applicable to flaws and leaks in metal structures and mitigation of corrosion damage were sought via literature review and discussion with vendors. In compiling a list of potential candidate technologies/methods, the description of need described above was not initially used to screen potential technologies. Therefore, techniques described as either temporary or permanent repairs were identified and put forth for consideration. Technical areas reviewed to obtain candidate technologies/methods included mitigation of chloride-induced stress corrosion cracking in dry storage cask systems for spent nuclear fuel, techniques identified for commercial tank/vessel repair, and emergency repair methods for ship/boat leak repair as well as pipeline repairs.

The evaluation group identified 26 separate technologies/methods as potential repair methods. These are listed in the second column of Table A-2, with a brief description of each technology provided in the third column. The 26 technologies were grouped into four categories and 12 high-level processes, which are indicated in the first column of Table A-2. The four categories consist of Coatings, Solid Phase Processing (SPP), Fusion Processing, and Non-process Repairs. The high-level processes associated with the four categories are as follows:

- Coatings – High-level processes consisting of gas, solution, liquid, solid-state, and non-metallic coating.
- Solid Phase Processing (SPP) – Includes the high-level processes of solid-state coating and FSW/P.
    - Note: Solid state coating is both a coating and an SPP technology that is identified as one or the other throughout the literature.
    - SPP methods are characterized by a lack of melting (for the substrate or additive material), which typically yields fine grain microstructures that contribute to superior mechanical properties compared to larger, less uniform grain structures.
- Fusion Processing – Due to the numerous processes and overlap among fusion process methods, this category was somewhat difficult to group into high-level processes. Many of the processes have been adapted from typical fusion joining processes (non-additive processes), powder feed,

and wire feed methods. The final high-level processes considered applicable by the evaluation team consist of laser welding, powder processing, arc welding (which includes numerous processes), and electron beam welding.

- As the category title indicates, all of these processes melt the material being deposited and some region of the substrate. This results in a deposit and surrounding area [i.e., heat affected zone (HAZ)] with a cast microstructure. Cast microstructures typically have material properties that are inferior to those resulting from solid phase processes. The melting of material also allows for changes in material composition and chemical interactions with contaminants, which further alter the material properties of the HAZ.

- Non-process Repairs – These technologies consist of no material bonding and were grouped into mechanical repair techniques (e.g., plugs and patches), which use compressive forces to create a seal, and methods using adhesion to connect materials and create a seal.

Table A-2. Identified candidate technologies/methods for infrastructure repair.

Category (High-Level Process)	Specific Technologies/ Methods	Description
Coating (gas state)	Physical vapor deposition (PVD)	Vacuum deposition (and high-temperature) method in which coating material goes from condensed phase to vapor phase and then forms a thin film as it condenses again. A solid precursor material is gasified, typically by using high-power electricity or laser. The gasified atoms are then moved into a reacting chamber where the coating substrate is located. Source material atoms then adhere to the substrate, forming a thin coat. Used to produce high-purity, specialized thin coatings. Thin films generated are typically created for functioning in high-stress environments. Considered a relatively high-cost, slow process and difficult to deploy remotely, which is limited to thin coatings.
	Chemical vapor deposition (CVD)	Similar to PVD in that it is used to produce high-purity, specialized thin coatings, CVD involves mixing the source material with one or more volatile precursors that function as a carrier device. The precursors, which are typically halides or hydrides, chemically interact and break down the source material. The entire CVD process is known to generate volatile by-products that must be safely removed via gas flow through the reaction chamber. Once created, the source material is transported by forced convection into the reaction chamber, which contains a substrate. Through the process of diffusion, reactants are deposited into the substrate. After the mixture adheres to the substrate, the precursor eventually breaks down, is removed by diffusion, and leaves behind the desired layer of source material on the substrate. The decomposition process can be facilitated or accelerated using heat, plasma, or various techniques. While CVD has limited applications, the process is considered a relatively economical deposition process compared to PVD.
Coating (solution state)	Plating – electroplating	Metal is deposited on a conductive surface and the coating “plates out” by electrodeposition onto the substrate/object being coated. An anode (positively charged) and cathode (negatively charged) are submerged in an electrolyte containing dissolved metal salts and ions needed to create a conductive solution. The anode may be soluble (made of the coating material) or insoluble. The cathode is the object/substrate to be coated. With a supplied direct current, dissolved metal ions in the electrolyte solution are reduced at the interface between the solution and the cathode.

Category (High-Level Process)	Specific Technologies/ Methods	Description
	Plating – electroless plating (chemical, auto catalytic plating)	Non-galvanic method consisting of several chemical reactions in an aqueous solution without the use of external electrical power to drive the reaction. The reaction results from hydrogen being released by a reducing agent (e.g., sodium hypophosphite). The hydrogen is released as a hydride ion, thus creating a negative charge on the substrate/object. This process is limited to thin coatings.
	Anodizing	An electrolytic process for creating oxide coatings. Most often applied to aluminum. The metal part to be treated (the anode) is submerged into an electrolytic solution bath along with a cathode. When a current is passed through the acid solution, hydrogen is released from the cathode and oxygen forms on the surface of the anode (part/substrate being coated), resulting in a thin metal oxide film growing on the surface. The buildup in coating thickness is limited for this process.
	Chemical conversion coating	Coating of metal surfaces via chemical or electro-chemical reactions/processes [e.g., chromate conversion (hexavalent chromium), bluing/black oxide coating]. Chemical conversion coating in general are not as robust against wear and corrosion as plated coatings and result in thinner layers.
Coating (liquid state – thermal spraying)	Flame spray	Additive process consisting of melted materials being sprayed onto a surface. Feed stock is heated via chemical means (i.e., combustion flame). Methods can provide coatings ranging from 20 µm to several millimeters. The process is thickness-limited and is not amenable for applying multiple layers through repeated processing. Thermal spraying can be used to apply metals, alloys, ceramics, plastics, and composites. Processes include both powder and wire form techniques. While the discharge temperature is high, the relative thermal energy flux to the surface during the process is not relatively high, which has allowed flammable surfaces to be coated. Particle velocities are relatively low, < 150 m/s.
	Arc spray	Similar to flame spray except feed stock is heated via electrical arc. Uses consumable metal wires that are charged and fed into a spray gun, resulting in an arc forming between the wires. As with flame spray, the process is thickness limited.
	Plasma spray	A thermal spray coating using a high-temperature (>15000 K) plasma jet generated by arc discharge. Higher temperature applications allow refractory materials such as oxide and molybdenum to be applied. Variations include atmospheric plasma spraying (APS), controlled atmosphere plasma spraying (CAPS), high-pressure plasma spraying (HPPS), low-pressure plasma spraying (LPPS), and vacuum plasma spraying (VPS). As with flame spray, the process is thickness-limited.
	Detonation spray (D-gun)	Highest-velocity method of the thermal sprays. Fuel mixture is detonated to create a shockwave of approximately 3500 m/s to propel coating materials. Process is capable of achieving porosities < 1% with relatively low oxygen contents between 0.1% and 0.5%. Process is used to apply metals and cermets (composite of ceramic and metals) and associated oxides of aluminum, copper, iron, and other metals. As with flame spray, the process is thickness-limited.

Category (High-Level Process)	Specific Technologies/ Methods	Description
	High-velocity oxy fuel	Thermal spray process using continuous combustion to propel additive material. Oxygen and fuel are fed into a combustion chamber. The resultant combustion produces pressures on the order of 1 MPa, resulting in supersonic discharge velocities. The feed stock powder is injected into the discharging flow. This process is often used for depositing cermet materials and metal alloys. As with flame spray, the process is thickness limited.
Coating (non-metallic powder coating)	Powder coating (nonmetallic coatings)	Powder coating metal objects accomplished by spraying the powder using an electrostatic gun (e.g., corona gun) or friction gun (e.g., tribo gun). The guns impart a positive electric charge to the powder, which is then sprayed toward the grounded substrate/object by mechanical or compressed air and then accelerated toward the workpiece by the powerful electrostatic charge. The object is then heated, and the powder melts into a uniform film and is then cooled to form a hard coating. Depending on the coating material, the substrate metal can be heated first and then the powder sprayed onto the hot substrate. The powder can also be applied using a fluidized bed in which the heated part being coated is submerged. Powder applications allow customized mixing of the powder and/or melt mixing of constituents and then generating new dry powder.
Coating (solid state) Solid Phase Processing	Cold spray also known as supersonic particle deposition	Additive manufacturing process relying on kinetic energy to deposit powder materials (metals, polymers, ceramics, composite materials, and nanocrystalline) via a heated inert gas. Applied at supersonic speeds up to 1200 m/s. The kinetic energy of particles causes softening (due to instantons heating) and plastic deformation upon impact, resulting in material bonding and a fine grain microstructure. Materials deposited remain below the melting point temperature (i.e., solid phase process). This process does not have a limit on coating thickness that can be achieved and allows the bonding of dissimilar materials. VRC Metal Systems LLC has demonstrated the mitigation of a small, active leak in a stainless-steel pipe pressurized to 60 psig.[2]
	Cold spray variants (subsonic process)	A lower-velocity (subsonic) variant of the cold spray process. Such processes use a sonic deposition nozzle in which particles are accelerated up to 1000 m/s. The lower-velocity application allows the process to be more efficient with carrier gas consumption. However, the lower velocity limits the range of applications because threshold energies required for material bonding cannot be obtained for many materials and material combinations. Forms of subsonic cold spray include low-pressure cold spray, kinetic cold spray, and Kinetic Metallization™.
Solid Phase Processing	Friction stir welding / processing (FSW/P)	A solid phase process that can be used for joining materials (friction stir welding) or the repair/mitigation of cracks or conditioning of sensitized zones (friction stir processing). FSW/P is effectuated by plunging a spinning tool into the workpiece. Heating due to friction and plastic deformation created by the rotation and axial force of the spinning tool result in a plasticized region, called the stir zone, below the shoulder of the tool. The FSW/P tool and associated stir zone are traversed across the joint or region to be reconditioned, resulting in a weld or processed region. Material temperatures resulting from FSW/P are typically 60% to 80% of the absolute melt temperature of the material being processed.

Category (High-Level Process)	Specific Technologies/ Methods	Description
Fusion Processing (laser welding)	Metal laser deposition / beam welding	Surface treatment process that is an additive process allowing material surfaces to be refurbished or built up in thickness. The laser melts a pool of metal on the workpiece and metal powder is deposited through a nozzle(s) into the pool, creating a new surface. Besides repairing or building up thickness, this process can be used to change surface properties.
	Laser beam welding (continues wave and pulsed laser)	Fusion welding process relying on a focused laser as the source of heat. The process is used for both metals and thermoplastics. Depending on how the laser is focused, the concentrated heat generated by the laser can allow for narrow, deep welds and high traverse rates. High-power density aids in reducing the size of the HAZ. Laser beam welding typically employs a shielding gas, can be automated, has a high production rate, and does not emit X-rays.
Fusion Processing (powder processing)	Powder bed process	Fusion process using powder to create three-dimensional objects or build up material on an existing substrate/object. The heat source used to melt and fuse the powder together can be a laser, electron beam, thermal energy, or fusion agent/energy. The process is applicable to metal and plastic parts.
	Metal powder coating	Process described under "Coating" category as the high-level process "non-metallic powder coating." For this application, only metallic-based coating is being considered. If a metal can hold an electromagnetic charge and withstand the elevated temperatures of the curing process, it can be powder coated.
Fusion Processing (arc welding – powder feed and wire additive manufacturing)	Arc welding (gas-metal arc welding, includes metal inert gas and metal active gas welding), gas-tungsten arc welding, flux core arc welding, shielded metal arc welding, etc.)	All these processes use similar methods of forming an electric arc between an electrode and the metal. This heats the metal and causes it to melt, creating a joint that fuses the metal pieces together. These processes can also be used to build up materials for cladding, repair, or additive manufacturing processes. These processes are capable of significant material buildup but result in HAZs due to material heating and fusion process and residual tensile stress resulting from shrinkage as the material cools.
Fusion Processing (EBW)	Electron beam welding (EBW)	Fusion welding process using a beam of high-velocity electrons focused at the interface of two objects to be joined. The kinetic energy of the electron beam is transformed into heat upon impact. The technology and associated control allow for very high surface power densities to be precisely focused on small areas. EBW has advantages for welding thin material or performing precision welds. Therefore, welds can be performed closer to heat-sensitive components. EBW can also be used to join dissimilar materials, where often only the material with the lower melting temperatures is melted. High-power density aids in reducing the size of the HAZ. Application is performed using a vacuum or partial vacuum environment. This process is typically performed in an autogenous mode (no filler material). For additive manufacturing or repair applications, the addition of a wire feeder is required.

Category (High-Level Process)	Specific Technologies/ Methods	Description
Non-Process Repair (mechanical repair techniques - plugs and patches)	Expanding plugs	Inserted plugs whose exteriors comprise the sealing surface that are then expanded once inserted through the penetration. These include expanding rubber screw plugs and pneumatically or hydraulically expanded plugs. Note: These repair methods are only applicable to full penetrations.
	Compression patches	Temporary plugs that consist of structural member(s) and compliant seal material that use mechanical pressure to compress the sealing material against the original substrate material without material bonds being created. Examples include: Inserted compression plate using structural member insert through leak to use interior surface of substrate to apply resulting force (i.e., sandwich substrate). Note: Only applicable to full penetration. • Exterior compression plate using external secondary surface to apply resultant force. This can include a secondary structure or the exterior surface of the substrate (e.g., banding, saddle clamps). Note: Applicable for mitigating a compromised/weakened location that does not leak. Insertable bladders – Repair consists of inserting bladder into tank/vessel to create liner. Uses interior surface of substrate to contain bladder/liner.
	Magnetic patches	For magnetic substrates, the magnetic force can be used to compress the sealing material. Magnetic patches can also be coupled with joining process to permanently attach the patch.
Non-Process Repair (adhesives)	Sealants	Consist of chemical mixture formulated to adhere to the substrate. Examples include silicone, tripolymer, rubber-based coatings, epoxy sealants and variants.
	Repair tapes - Silicon, silica, and silicone, and copolymer hybrid tapes and variants	Repair tapes applicable for adhesion on rough surfaces and harsh environments. Examples include: Thermo-Trex® high-temperature silica tape – Made of 96% pure SiO_2 silica fiber, which offers superior resistance to radiant heat and flame. Suitable for continuous use at 1800°F and will withstand short-term exposure with temperatures as high as 2300°F. Permatex silicone fusion tape – Made of specially formulated silicone rubber and contains no adhesives. It chemically bonds to itself upon contact. Used extensively by the military for field repairs.
	Carbon fiber wraps and variants	A carbon fiber vinyl (usually a PVC-based vinyl) having an adhesive backing.

Preliminary Screening / Down-Selection of High-Level Processes

The down-select process focused on the high-level processes, with the individual technologies/methods identified serving as examples associated with each high-level process.

For the 12 high-level processes listed in Table A-2, a preliminary screening was conducted to determine if any of the high-level processes should be eliminated from the head-to-head comparison. The following criteria were put forth as guidelines for the initial screening:

- Repairs are to be considered long term and semi-permanent (lasting 40 years).
    - Note: No repair was considered permanent due to the potential impacts of the corrosive and radioactive environment. To assign a designation of permanent repair would require further assessment.

- Repairs are not to rely on non-metallic materials that have not undergone dose testing to assess service life in the radioactive environment.

Based on these two criteria, the following processes were eliminated from further consideration: the category “Non-Process Repair,” consisting of the high-level processes of mechanical and adhesive methods, and the high-level process “non-metallic powder coatings,” from the “Coating” category. These three high-level processes were excluded due to the use of non-metallic materials in the repair for which application to radioactive environments was not currently known and further investigation was considered beyond the scope of this initial down-select process.

In considering the high-level process “gas state coating,” from the “Coating” category, the evaluation group deemed it impractical due to complexities associated with setting up and conducting the process for remote application and eliminated the process from further consideration.

Therefore, the preliminary screening reduced the number of high-level processes to be carried forward for a head-to-head comparison from the original 12 to the following 8:

- Solution-state coating
- Liquid-state coating (thermal spraying)
- Solid-state coating (considered both a coating and a solid phase process)
- Solid phase processing (friction stir welding)
- Laser welding – fusion process
- Powder processing – fusion process
- Arc welding – fusion process
- Electron beam welding – fusion process

Comparative Evaluation

The methodology used for the comparative evaluation of the eight candidate high-level processes was similar to previous technology evaluations conducted for the mitigation and repair of chloride-induced stress corrosion cracking in dry cask storage systems for spent nuclear fuel.[3,4] Based on the need description formulated by PNNL staff (see above), previous comparative evaluations, and past experience of the evaluation group members, seven high-level attributes specific to this application were identified for consideration. Factors associated with each attribute were identified by the evaluation group. These attribute factors are the individual criteria used for rating the candidate technologies/methods. Table A-3 lists the seven high-level attributes selected for the evaluation along with their associated attribute factors.

A subjective weighting was assigned to each attribute based on the anticipated priority of the attribute as applied to this application. The higher the percentage assigned to the attribute, the greater the importance (i.e., weighting) given to that attribute. The methodology applied used a percentage weighting such that a perfect score for a technology would equal 100.

The assigned weightings for each high-level attribute are listed in Table A-3 and are based on the subjective scale provided in Table A-4. “Operational suitability” and “properties of deposit” were

[3] Tatman J. 2018. *Welding and Repair Technology Center: Extended Storage collaboration program Canister Mitigation and Repair Subcommittee – Industry Progress Report*. Electric Power Research Institute, Palo Alto, CA.
[4] Ross K and M Alabi. 2019. *Update on Investigations of Viability of Cold Spray and FSW as a Spent Nuclear Fuel Dry Storage Canister Mitigation Tool*. SFWD-SFWST-M3SF-19PN010201086, PNNL-29217, prepared for the U.S. Department of Energy by Pacific Northwest National Laboratory, Richland, WA.

identified by the evaluation group to be of highest priority and each was given a weighting of 20% of the final score, (i.e., “Very important” per Table A-4). While the “ability to deploy remotely” and “verification of the repair” were considered high-priority items, they were weighted less, 15%, as engineering development could potentially improve these attributes for this specific application. In comparison, the “operational suitability” and “properties of deposit attributes” are considered related to the physics and process methodology, regardless of the specific application. The “substrate impact” was considered similar in importance to the “ability to deploy remotely” and “verification of the repair” and was thus also assigned a weighting of 15% of the final score. Due to the nonproduction, non-commercialization, and limited application aspects of the repair process, “economics” was viewed as the least important attribute and was assigned a weighting of 5% of the total score. Because of the unique application of the repair technology and limited applications in the nuclear waste environment, the attribute of “technology accessibility” was given a weighting of 10%, which is less than all others except “economics.”

The individual attribute factors were scored based on a scale of 0 to 5. The subjective scale applied for scoring the individual attribute factors is also presented in Table A-3. For each attribute and candidate technology, the scores for the associated attribute factors were summed and a relative score calculated based on the percent weighting for the individual attribute. However, if a score of 0 was given a single attribute factor, the associated attribute was given a score of zero for that technology. For example, the attribute “Substrate impacts” has five attribute factors listed in column three of Table A-4. If each attribute was given a score of 5, the sum of the attribute factors would be 25 (i.e., a perfect score), but the relative attribute score would be 15 (i.e., equal to the weighting percent of the attribute, refer to column 4 of Table A-5). Considering the same attribute, if each attribute factor was given a score of 3, the sum of the attribute factors would be 15, but the relative attribute score would be 9. However, if any single attribute factor was given a score of 0, then the relative attribute score would default to 0.

The scoring of the technologies was conducted independently by each member of the evaluation group. The attribute scores applied by each evaluation group member were whole numbers, 0, 1, 2, 3, 4, or 5 (i.e., no decimal values used). The average of the individual relative attribute scores was calculated from the individual relative attribute scores of evaluation group members for each technology and these scores are presented in Table A-1. The total scores presented in Table A-1 are the sum of the averaged relative attribute scores.

Table A-3. Attribute groups, factors, and weighting for conducting comparative evaluation of candidate repair technologies.

High-Level Attribute	Attribute Factors	Weighting of High-Level Attribute (% of ranking)
Ability to deploy remotely	- Size of deployable equipment - Portability of equipment - Application to remote deployment - Application in hazardous environment - In situ process	15
Operational suitability	- Ability to automatically change/replenish consumables - Control of operational parameters - Process temperature at substrate - Impact of gravity / repair orientation - Process deposition rate - Process stability/sensitivity	20

High-Level Attribute	Attribute Factors	Weighting of High-Level Attribute (% of ranking)
Properties of deposit	- Deposit density / seal ability - Achievable deposit thickness - Uniformity / variability of deposit / deposit properties - Susceptibility to corrosion - Ability for deposit to match substrate properties	20
Substrate impacts	- Pretreatment requirements - Process impacts on substrate - Post-process residual stress state - Potential distortion of substrate - Post-treatment required	15
Verification	- Ease of repair inspection - Ability to verify repair integrity - Ability to distinguish between acceptable and unacceptable repairs	15
Technology accessibility	- Commercial availability - Maturity of technology - Industry experience	10
Economics	- Capital cost - Operating cost, including consumables and utilities	5

Table A-4. Subjective scales used for weighting attributes and scoring attribute factors.

Attribute scoring	Renders process impossible	Highly dis-advantageous	Somewhat dis-advantageous	Neither advantage nor disadvantage, or undetermined	Somewhat advantageous	Highly advantageous
	0	1	2	3	4	5
Attribute category weight		Unimportant	Somewhat important	Moderately important	Very important	Critical
	Low %	0	6	11	16	21
	High %	5	10	15	20	25

Summary

For the down-select process, the attribute factors could be expanded or modified to tailor the comparative evaluation to a more specific application or environment. The scoring of the attribute factors is subjective but provides a relative comparison of the technologies for each single attribute. The weighting of the attributes to obtain a final score is subjective and needs to be evaluated by the final end users. Using the same attribute factor scores, a different ranking of the technologies may be obtained based on weighting/prioritizing the attributes differently. For example, Table A-1 shows that the attribute "ability to deploy remotely" is weighted at 15 out of 100. A perfect score from the team (all three members rating all attribute factors a 5) would be 15. The relative attribute scores range from 14.2 to 3.8, indicating that the team views some high-level processes as easier to deploy remotely than others. The choice of weighting is important because the total scores range from 76.6 to 40.9 out of 100, so there were no perfect scores. The PNNL team agreed that the attribute weights used in the evaluation were appropriate for infrastructure repair for the EM mission but recognizes that differences in priorities (changing the weight of each attribute) could change the order in the overall ranking of the high-level processes.

This letter is intended to present a down-selection process for comparing potential repair technologies and to provide a basis for why cold spray was put forth as a potential candidate repair system to be evaluated for conducting infrastructure life-extension repair relative to corrosion and wear for the EM mission.

Sincerely,

Carl Enderlin
Chief Experimentalist
Computational and Experimental Engineering Group
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Appendix B

Repair Task Group Technology Summary Tables

The repair task group operates on a consensus basis and consists of industry subject matter experts whose efforts are supported by their employers. One objective of the repair test group is to identify and evaluate methods/ practices (tools) to repair environmental degradation of DCSS canisters. Pursuant this goal the task group held a series of meetings and conference calls to establish needs, develop understanding, generate concepts, and score concepts/methods for repair of canisters. The results are recorded in technology evaluation tables shown in Appendix B. Table B-1 through Table B-3 show that scoring differs for repair within an overpack, with overpack removed and considering fast confinement only. Learnings from this effort assisted in down selection of technologies of interest (Tatman 2018) to a smaller set of technologies for further investigation.

These consensus-based efforts rely on the existing knowledge, expertise, and judgment of those who participate. The task group included owner/operators, cask vendors, engineers, and technology experts.

The charts provided in this section graphically presents an informal consensus-based evaluation based on knowledge of repair task group participants and does not represent results of a formal study. They are provided as examples of effective low effort screening tool for technology evaluation.

Table B-1. **<u>Within the overpack</u> *draft* technology summary sheet for canister repair technologies compiled by the EPRI ESCP Repair Task group and presented by Ken Ross at the Fall EPRI ESCP Meeting 2019. This chart graphically presents an informal consensus-based evaluation based on knowledge of repair task group participants and does not represent results of a formal study. It is being provided as an example of an effective low effort screening tool for technology evaluation.**

Repair Technique	Operating Experience		Feasibility / Accessibility	Effectiveness Against SCC		No Impact on Inspectability	Type of Repair (Interim / Permanent)	Commercial Availability	Comments
	ISFSI	Other		Repair Area	No Detrimental Effect				
Fusion Processes									
Arc Welding	High	High	Med	Low	Low	High	Both	High	High heat input sensitizes surrounding base material.
Arc-Welded Overlay / Patch	High	High	Med	Low	Low	High	Both	Low	High heat input sensitizes surrounding material. Added material increases risk of accessibility.
Arc-Brazing / Soldering	Low	High	Med	High	High	High	Interim	Low	Can be done at sufficiently low temperatures to avoid sensitization. Comparatively reduced repair strength.
Thermal Spray	Low	High	Med	Med	Med	Med	Interim	Med	High heat input sensitizes surrounding material.
Solid Phase Processes									
Friction Stir Welding	Low	Med	Low	High	High	High	Permanent	Low	Top performer for strength of repair. High technical risk for within overpack, low technical risk with overpack removed
Cold Spray	Low	High	High	High	High	High	Both	Med	Fast repair, easy to inspect and does not damage surrounding material.
Peening	Med	High	Med	High	High	High	Interim	High	In current use at SONGS.
Non-Metallics									
Coatings / Sealants	Low	High	Med	Med	High	Med	Interim	Low	Will need to be further categorized.
Silicon Hybrid Copolymer Tape (Keeno)	Low	Med	Med	High	High	Low	Interim	Low	EDF has installed this material at several plants in France, SNF transfer canals.
Carbon Fiber Wraps	Low	Med	Med	High	High	Low	Interim	Low	ASME to address / codify installation requirements.

Table B-2. **<u>Overpack removed</u> *draft* technology summary sheet for canister repair technologies compiled by the EPRI ESCP Repair Task group and presented by Ken Ross at the Fall EPRI ESCP Meeting 2019. Evaluation for technolgies to repair casks outside of an overpack. This chart graphically presents an informal consensus-based evaluation based on knowledge of repair task group participants and does not represent results of a formal study. It is being provided as an example of an effective low effort screening tool for technology evaluation.**

Repair Technique	Operating Experience		Feasibility / Accessibility	Effectiveness Against SCC		No Impact on Inspectability	Type of Repair (Interim / Permanent)	Commercial Availability	Comments
	ISFSI	Other		Repair Area	No Detrimental Effect				
Fusion Processes									
Arc Welding	High	High	High	Low	Low	High	Both	High	High heat input sensitizes surrounding base material.
Arc-Welded Overlay / Patch	High	High	High	Low	Low	High	Both	High	High heat input sensitizes surrounding material. Added material increases risk of accessibility.
Arc-Brazing / Soldering	Low	High	High	High	High	High	Interim	High	Can be done at sufficiently low temperatures to avoid sensitization. Comparatively reduced repair strength.
Thermal Spray	Low	High	High	Med	Med	Med	Interim	High	High heat input sensitizes surrounding material.
Solid Phase Processes									
Friction Stir Welding	Low	Med	High	High	High	High	Permanent	Med	Top performer for strength of repair. High technical risk for within overpack, low technical risk with overpack removed
Cold Spray	Low	High	High	High	High	High	Both	Med	Fast repair, easy to inspect and does not damage surrounding material.
Peening	Med	High	High	High	High	High	Interim	High	In current use at SONGS.
Non-Metallics									
Coatings / Sealants	Low	High	High	Med	High	Med	Interim	High	Will need to be further categorized.
Silicon Hybrid Copolymer Tape (Keeno)	Low	Med	High	High	High	Low	Interim	High	EDF has installed this material at several plants in France, SNF transfer canals.
Carbon Fiber Wraps	Low	Med	High	High	High	Low	Interim	High	ASME to address / codify installation requirements.

Table B-3. **<u>Fast confinement</u> *draft* technology summary sheet for canister repair technologies compiled by the EPRI ESCP Repair Task group and presented by Ken Ross at the Fall EPRI ESCP Meeting 2019. Evaluation for technolgies to execute fast confinemet an identied leak of a canister. This chart graphically presents an informal consensus-based evaluation based on knowledge of repair task group participants and does not represent results of a formal study. It is being provided as an example of an effective low effort screening tool for technology evaluation.**

Repair Technique	Operating Experience		Feasibility / Accessibility		Effectiveness Against SCC		No Impact on Inspectability	Type of Repair (Interim / Permanent)	Commercial Availability		Comment
	ISFSI	Other	In-Situ	Ex-Situ	Repair Area	No Detrimental Effect			In-Situ	Ex-Situ	
Fusion Processes											
Arc Welding	High	High	Med	High	Low	Low	High	Both	High	High	High heat input sensitizes surrounding base material.
Arc-Welded Overlay / Patch	High	High	Med	High	Low	Low	High	Both	Low	High	High heat input sensitizes surrounding material. Added material increases risk of accessibility.
Arc-Brazing / Soldering	Low	High	Med	High	High	High	High	Interim	Low	High	Can be done at sufficiently low temperatures to avoid sensitization. Comparatively reduced repair strength.
Thermal Spray	Low	High	Med	Med	Med	Med	Med	Interim	Med	High	High heat input sensitizes surrounding material.
Solid Phase Processes											
Friction Stir Welding	Low	Med	Low	High	High	High	High	Permanent	Low	Med	Top performer for strength and repair. High technical risk for within overpack, low technical risk with overpack removed
Cold Spray	Low	High	High	High	High	High	High	Both	Med	Med	Fast repair, easy to inspect and does not damage surrounding material.
Peening	Med	High	Med	High	High	High	High	Interim	High	High	In current use at SONGS.
Non-Metallics											
Coatings / Sealants	Low	High	Med	High	Med	High	Med	Interim	Low	High	Will need to be further categorized.
Silicon Hybrid Copolymer Tape (Keeno)	Low	Med	Med	High	High	High	Low	Interim	Low	High	EDF has installed this material at several plants in France, SNF transfer canals.
Carbon Fiber Wraps	Low	Med	Med	High	High	High	Low	Interim	Low	High	ASME to address / codify installation requirements.