

COLD SPRAY COPPER COATING APPLICATION TO THE WELD CLOSURE ZONE OF USED NUCLEAR FUEL CONTAINERS

Peter Keech¹, Phuong Vo², Dominique Poirier², Jean-Gabriel Legoux², Jason D. Giallonardo¹, David Doyle¹

¹ Nuclear Waste Management Organization, Toronto, Ontario, Canada
(pkeech@nwmo.ca)

² National Research Council, Boucherville QC, Canada

Abstract

Since 2011, NWMO has been developing copper coatings for the purpose of providing corrosion protection to steel used nuclear fuel containers. Through a series of initiatives in cold spray, electrodeposition and weld cladding methods, copper coatings ranging in thicknesses of 3 to 10 mm have been applied to a series of steel substrates. Components have progressed from small coupons to full sized parts. NWMO has determined that both electrodeposition and cold spray processes will be used to copper coat the UFC. Electrodeposition will be used for copper coating the “factory” supplied UFC components (shell, lid). This process is a well-established, straight-forward industrial technique well suited to the UFC geometry under conventional fabrication conditions. The cold spray process is to be used for coating the closure weld upon final assembly; this process is well suited to automated, robotic application in a radioactive environment. In the most recent investigation, cold spray copper has been applied across mock-up and actual weld zones to complete the coating of used fuel containers. Within this work, cold spray coating parameters must be sufficiently rugged to produce high quality coatings across a region including electrodeposited copper on both edges (transition zone), container steel and welded steel.

1. Introduction

In 2002, the Nuclear Waste Management Organization (NWMO) was established in accordance with the Nuclear Fuel Waste Act to assume responsibility for the long-term management of Canada's used nuclear fuel. Through an extensive dialogue with Canadians, the NWMO developed a comprehensive plan to manage the used nuclear fuel by:

- 1) Working with the nuclear operators to retrieve it from reactor sites' interim storage;
- 2) Transporting it to repackaging facilities;
- 3) Sealing it in within extremely long lived containers; and
- 4) Emplacing it into a Deep Geological Repository (DGR).

Figure 1 illustrates the current conceptual approach for the long term storage of radioactive fuel bundles in specially designed containers for CANDU fuel. The containers would be emplaced in the rock of a suitable geological formation at a minimum of 500 m of depth, for an intended lifespan of more than 100,000 years (i.e., permanent storage). The Used Fuel Container (UFC) is

a major engineered component of the design of the DGR. NWMO's UFC design is a mid-sized capacity vessel (12 bundle/layer x 4 layers = 48 bundle), which incorporates a carbon steel core for structural strength to withstand glacial loading and a 3 mm thick exterior copper coating for corrosion resistance. The 3mm coating thickness is based on very conservative assumptions for corrosion in a DGR environment, to which less than 1.3 mm copper loss is expected in one million years [1,2];

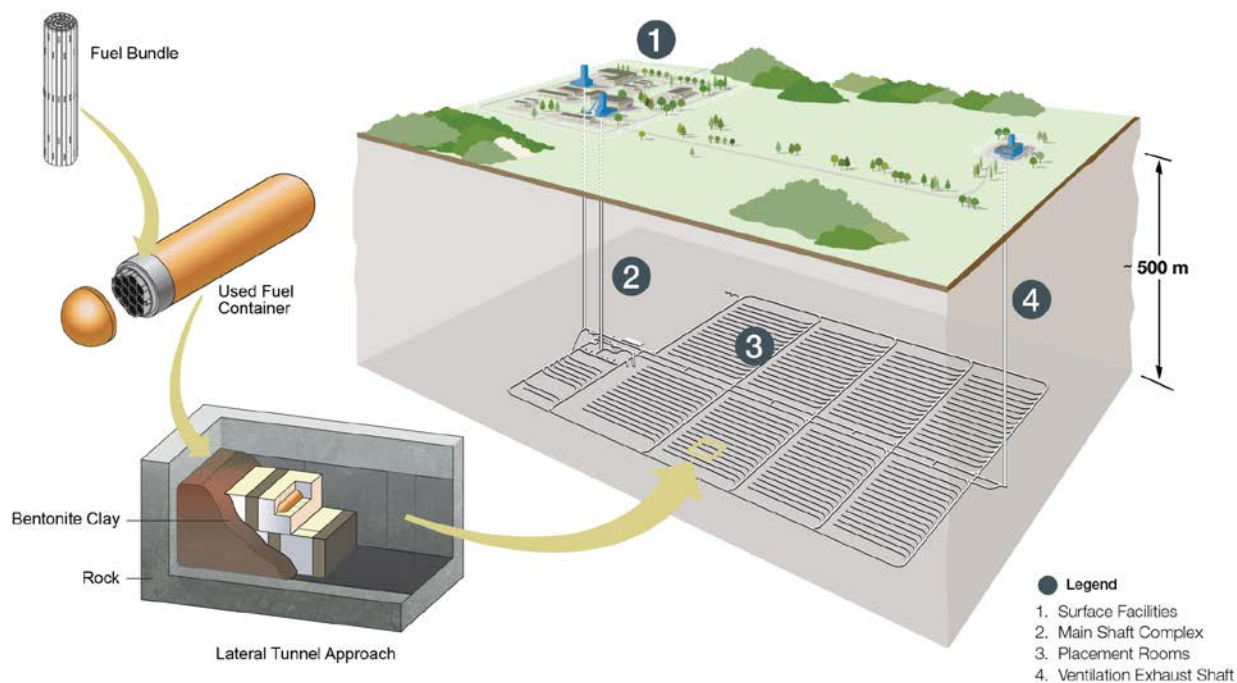


Figure 1: View of deep geological repository

The development of copper coatings for application to UFCs has been ongoing for several years by the NWMO and its research partners. From a list of coating/weld cladding technologies, two front runner methodologies emerged: electrodeposition[3], and cold spray[4]. Among coating methods, the former technology offers a substantial inherent advantage, in that it has been widely used for many applications over a wide range of industries, including nuclear[5], to produce pure, coherent metallic coatings, with exceptional adhesivestrength to the substrate. However, electrodeposition does require modest to significant fabrication times, as the layer-by-layer deposition of the metallic copper is limited mainly by the applied current density based on the given electrolyte. For coatings such as those required for UFCs, days may be required to achieve the full thickness via electrodeposition. This duration is quite acceptable for UFCs that have not been filled with used nuclear waste (i.e., at a container fabrication facility); accordingly, electrodeposition will be utilized to coat copper onto empty containers and lids. These components will be delivered to the used fuel packing plant (UFPP) in preparation for fuel loading.

At the UFPP, the containers will be filled with spent fuel and welded shut via the laser welding methodology[6]. At this point, the corrosion barrier will be completed by applying copper to the closure weld region using the cold spray methodology in the radioactive UFPP. Because this solid-state coating method allows for very rapid coatings to be applied from powders onto substrates, without changing chemistry/oxidation state of the powder material[7], it is ideal for this environment. However, its usage is not as wide-spread industrially as electrodeposition. In consideration of this, the NWMO has spent additional efforts on assessing the coating properties[8] prior to utilizing this method for UFCs. To bridge this technology gap, the most recent efforts have been directed at blending the electrodeposited coating to the cold spray coating in trials, so that a viable UFC production method can be obtained.

2. Cold Spray Process

2.1 Cold Spray Theory

As described extensively elsewhere[9], cold spray is a solid state process that uses a high-speed gas to send powder toward a substrate where it deforms and strongly adheres; a schematic of the process is presented as Figure 2[10]. The De Laval nozzle allows for gas to accelerate to a supersonic rate and obtain the speeds necessary to produce the well-adhered coating.

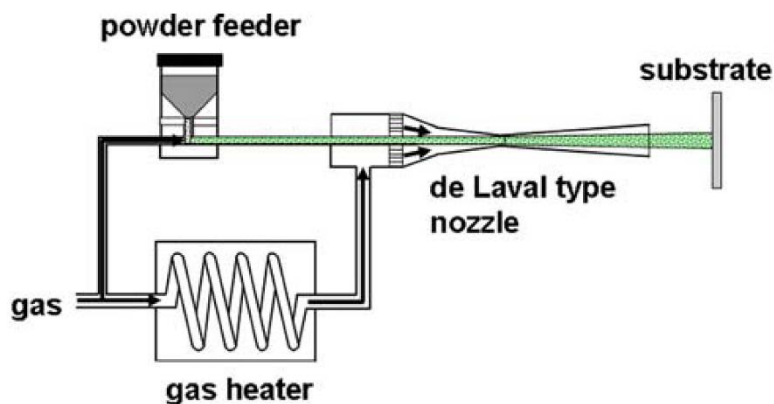


Figure 2: Schematic diagram of cold spray process

2.2 Copper Cold Spray Parameters for UFCs

To date, coatings have been produced using Plasma Giken PCS-800 or PCS-1000 machines, located at the National Research Council (NRC) facility in Boucherville, QC. The former has proven to be less susceptible to powder clogging. Both of these devices can be classified as “high pressure” cold spray devices, for which significant ongoing development is occurring, in the research and manufacturing communities. In addition to the results and methods presented within this work, there is an ongoing parallel effort to broaden the cold spray and associated surface preparation techniques. At this time, the previous work has resulted in a reference cold spray process[11].

To produce suitable coatings for UFCs, it is presently necessary to utilize helium as a carrier gas for an initial adherent layer; otherwise, the velocity of the copper particles is insufficient to

strongly adhere the copper to the steel. Subsequent cold spray passes can be made using nitrogen as a carrier gas. Independent of the gas type, the other conditions used to produce the coatings are:

- 1) Steel substrate (A516 Grade 70 or equivalent), with a minimum thickness of 25 mm, grit blasted using 24 grit alumina;
- 2) Pressure of 4.9 MPa;
- 3) Gas Temperature 800°C;
- 4) Feed rate between 20 and 200 g/min;
- 5) Standoff distance of 30 mm.

Upon completion of the coating deposition, it is necessary to machine the surface to allow for ultrasonic inspection. In addition to this, the coating must be heat treated to impart ductility; prior to heat treatment, elongation as measured via ASTM E8, is typically less than 1%. After a heat treatment at 350°C for 1 hr, the copper ductility is restored, such that an ASTM E8 measurement will exceed 20% elongation prior to fracture.

Following the previous phases of work, the NWMO and NRC initiated the development of a “Cold Spray Manual”, a document package that will consist of cold spray specifications and analysis methods. As this document package matures, it will undergo extensive validation testing and review, and will be publicly available. Prior to this event, it must incorporate work that demonstrates the blending of the electrodeposited and cold spray coatings, the first of which is described below.

3. Cold Spray Production Results

The development sequence of part production has been following the typical production schedule of the NWMO-NRC collaboration:

- 1) Production of coatings on small, planar parts/coupons;
 - o Extensive characterization of parts, including destructive testing;
 - o Development of feedback to the spray conditions and modification of parameters, as required;
- 2) Production of modest sized cylindrical parts;
 - o Characterization (as above)
 - o Feedback to production (as above)
- 3) Production of full-scale parts;
 - o Characterization (as above)
 - o Feedback to production (as above)
- 4) Update of Cold Spray Procedures and Operations and the Cold Spray Manual.

Efforts to date have included items mentioned in 1) and 2), i.e., planar and cylindrical parts; while ongoing work will focus on full-scale parts and documentation updates.

As noted above, the initial tests consisted of deposition of cold spray coatings onto planar samples;

Figure 3 illustrates before and after views of a typical planar coupon.

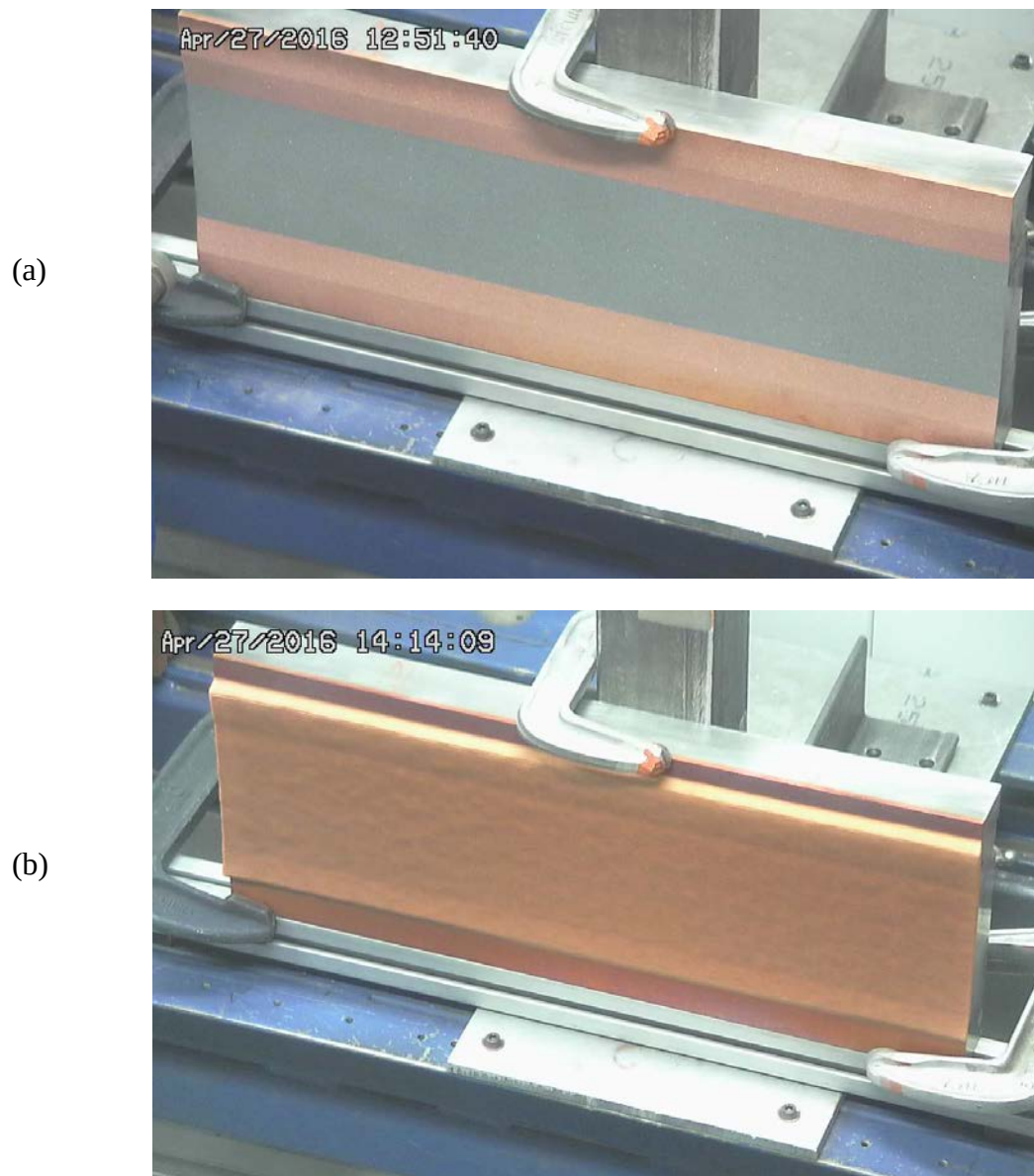


Figure 3: Cold spray development components illustrating (a) a planar coupon with only electrodeposited material at the edges and; (b) the cold spray zone completely covered.

As can be observed in Figure 3, it is necessary to overspray the electrodeposited zone with cold sprayed copper; this is because the electrodeposited zone is machined by tapering it back to the bare steel at a 10° angle.

Following production, the samples were annealed, and assessed for adhesion, via ASTM C633 methods. As this method uses a cylindrical puck machined from the coating, the adhesion measurement spans the region where cold sprayed copper is applied to the bare steel, as well as the region where it is applied to the electrodeposited copper. The measured values for adhesion

were very high; all exceeded the minimum specified value of 60 MPa at which the adhesive fails. From these results, it became clear that spray parameters did not need adjustment prior to spraying on cylindrical parts. However, it is also clear from looking at the above image that there is a significant shoulder at the upper and lower edges of the cold spray zones, where the coating is much thicker than the underlying electrodeposited regions. To eliminate the formation of this shoulder, and minimize the requirement for annealing, it is necessary to adjust the programming of the robot to spray these zones less in the centre of the cold spray zone. This is a topic for future work.

Following the trials on planar coupons subsequent efforts were made to produce coatings on parts that mimicked the size and shape of the container: short rings. A typical effort is illustrated in Figure 4.

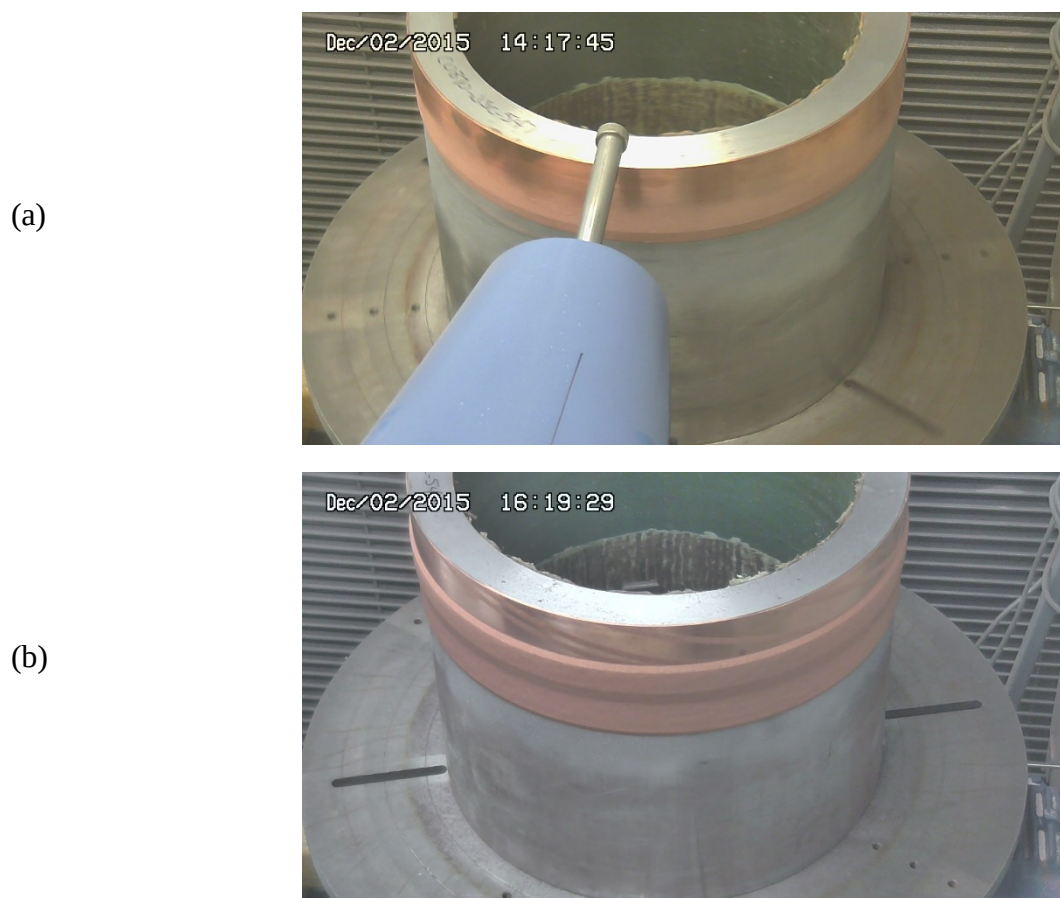


Figure 4: Cold spray development components illustrating: (a) a cylinder with only electrodeposited material at the edge and; (b) coverage by cold spray to produce the blended zone

The above work was then followed by the application of cold spray coatings to a simulated weld zone, where electrodeposited coatings border the transition zone, as shown in Figure 5. For the simulated weld zone, the substrate consisted of two pipe sections that had been welded together via laser welding (instead of a hemispherical head welded to a pipe section). Thus, the cold spray coating was applied to welded steel, the heat affected zone, unaffected steel and the electrodeposited copper zones.

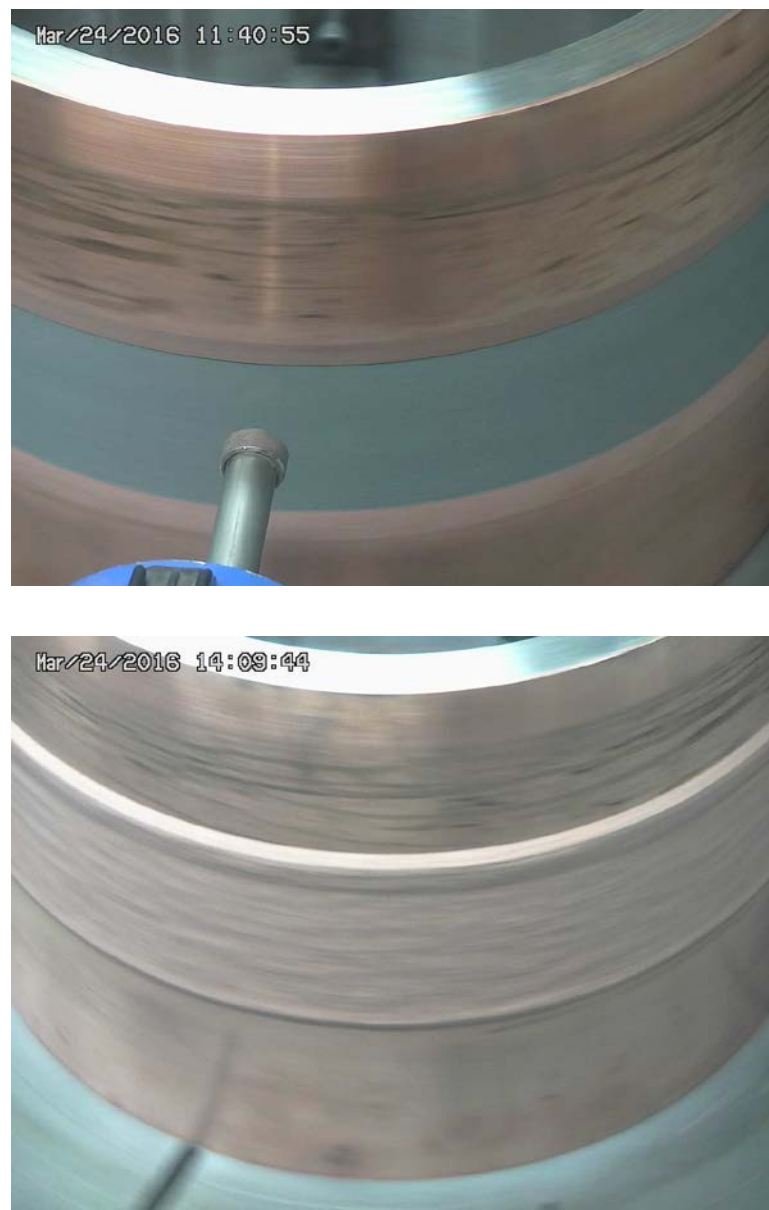


Figure 5: Cold spray development components illustrating (a) a cylinder with only electrodeposited material at the edges around a simulated weld zone and; (b) coverage by cold spray to produce the blended zone across the weld zone.

The coatings were characterized by metallographic analysis, much of which is still ongoing, and will be presented in the future. Adhesion results were obtained similar to the planar coupons: by applying a pulling force to the coating away from a substrate. While adhesion is consistently strong in that the coating does not fail prior to the failure of the adhesive used in the ASTM-C633 test, these results are not truly quantitative, owing to the curvature of the substrate. In this case, delamination is complicated by shear forces, as the substrate is not perfectly perpendicular to the delaminating force, particularly at the edges of the test coupons.

The “semi-quantitative” character of the pull tests has been used by the program as feedback to the order of the operations, post deposition of the cold spray coating. In some cases, samples were machined following cold spray, then subjected to heat treatment, while other samples were heat treated, and then machined. Adhesion measurements revealed that there was no discernable difference among samples: they all achieved the minimum adhesion strength of 60 MPa. This result has significance to the future, in that within a used fuel packing plant (UFPP), the order of operations will produce changes to the robotic systems that will be used to manipulate the UFCs. The demonstration of the flexibility of the cold spray and related processes will allow systems designers to optimize the UFPP for other important features, such as safety and speed, rather than for individual steps within the cold spray process.

The most recent and ongoing efforts within the testing program include demonstration of the cold spray coating onto the full sized containers. While a prototype container has been sprayed over the weld zone with cold spray, the coating edges to which it blended were also cold spray, which had previously been used to coat an entire container during product development (Figure 6).



Figure 6: Prototype cold-spray coated container coated on the weld zone using cold spray.

It is expected that within several months, a series of electrodeposited containers will be similarly coated with cold spray copper in the weld zone.

4. Conclusions and Future Work

The successful application of the cold spray coating technology to simulated weld closure zones was preliminarily demonstrated through activities involving:

- 1) Coating initial development for planar samples;
- 2) Coating development on cylindrical samples;
- 3) Extensive material characterization coating; and

- 4) A single effort on a cold-spray vessel with an actual weld zone.

Further development of the cold spray process for the coating of the weld zone within a clear operating window is currently underway.

5. References

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