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Characterization of commercially cold sprayed copper coatings and determination of the effects of impacting copper powder velocities



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ABSTRACT

Copper coated steel containers are being developed for the disposal of high level nuclear waste using processes such as cold spray and electrodeposition. Electron Back-Scatter Diffraction has been used to determine the microstructural properties and the quality of the steel-copper coating interface. The influence of the nature of the cold-spray carrier gas as well as its temperature and pressure (velocity) on the coating's plastic strain and recrystallization behaviour have been investigated, and one commercially-produced electrodeposited coating characterized. The quality of the coatings was assessed using the coincident site lattice model to analyse the properties of the grain boundaries. For cold spray coatings the grain size and number of coincident site lattice grain boundaries increased, and plastic strain decreased, with carrier gas velocity. In all cases annealing improved the quality of the coatings by increasing texture and coincidence site-lattices, but also increased the number of physical voids, especially when a low temperature cold spray carrier gas was used. Comparatively, the average grain size and number of coincident site-lattices was considerably larger for the strongly textured electrodeposited coating. Tensile testing showed the electrodeposited coating was much more strongly adherent to the steel substrate.

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1. Introduction

The currently accepted approach to the long-term management of Canada's used nuclear fuel is Adaptive Phase Management (APM), which includes the option of sealing the used nuclear fuel in metal containers emplaced in a suitably selected deep geological repository (DGR) [1]. The current container design consists of a steel vessel (Canada) or cast iron insert (Sweden, Finland) with an external copper (Cu) shell which acts as a corrosion barrier. Following an early brief emplacement period, during which the containers will be warm due to radiation decay within the spent fuel, and oxygen trapped during emplacement activities will cause some corrosion, the repository conditions will be cool and humid or wet and anaerobic [2]. As a consequence, Cu is an ideal container material since it is thermodynamically stable under these

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anaerobic conditions [3]. Many archaeological (i.e., anthropogenic) and natural (i.e., natively formed metallic) artefacts confirm this stability [4,5].

To date, the majority of Cu container research has been on wrought Cu, of the form that can be readily manufactured into cylindrical components via extrusion, pierce and draw, and rollforming methodologies [6–9]. However, for a dual-walled Custeel/cast iron container, ongoing manufacturing challenges exist, despite the fabrication of prototypes by Posiva (Finland) [10,11] and SKB (Sweden) [12]. To achieve sufficient structural support and ideal (i.e., round and non-ovalic) shape, both during and after manufacturing, cylindrical components must be significantly oversized and machined down extensively. An approximate additional 40% of the finished container weight must be removed and recycled in this process, even for a finished product that has a 5 cm final wall thickness. A further complication is the need to avoid long-term creep after emplacement within the DGR which requires tight machined tolerances between the inner vessel and outer shell; i.e., a nominal gap of <1.5 mm over the full container length (>4 m). To overcome these issues, Cu coated steel containers are being investigated by the Nuclear Waste Management Organization (NWMO, Toronto Ontario, Canada). The coatings would be formed directly on the surface of the steel vessel with a thickness defined by the need for corrosion protection. Recent calculations [13,14] suggest a corrosion allowance of only 1.27 mm would be required to prevent failure by corrosion under the conditions anticipated in a DGR.

The use of commercial processes, such as cold spray coating and electrodeposition are being considered [15,16]. Cold spray coating is a material deposition process that accelerates small (approximately 10–100 μ m in diameter) un-melted powders to high velocities (500–1200 m/s) using a supersonic jet gas stream [17,18]. The low applied temperatures and short applied time scales limit oxidation and changes in the physical properties of the feedstock particles making cold spray coating an attractive option compared to other commonly used thermal spraying techniques. However, as produced, the resultant Cu coating is expected to have significant residual cold work, with additional hardness and a low fracture toughness. These properties can be altered to produce a more conventional form of Cu by post-deposition annealing at moderately low temperatures (i.e., 300–400 °C) [19].

To justify the adoption of such a technology for used-fuel containers the microstructural features of the coatings and the quality of the interface between the Cu and the steel vessel must be evaluated. Electron Back-Scattered Diffraction (EBSD)—a technique that has been adopted as a standard for determining grain sizes [20]and electron microscope imaging techniques have been applied to determine grain size, orientation and texture, as well as the distribution of strain within the coatings and across the Cu/steel interface. This study is part of an ongoing program to evaluate the viability of different coating processes [15,21,22] by determining the most cost-efficient coating parameters (i.e., spraying temperature, pressure and carrier gas), and their mechanical properties and degradation (corrosion) rates. The application of EBSD will help to optimize the choice of coating parameters required to satisfy mechanical design requirements and to identify the microstructural features which could be important in determining corrosion rates. A detailed study of the corrosion performance of these coatings are published elsewhere [23].

2. Experimental

2.1. Cold spray Cu feedstock powders

Commercially available Cu feedstock powders (Inovati,



Fig. 1. Photograph of a typical Cu coating cross-section used for microscopy analyses.



Fig. 2. Modified test jig used to mount and apply tensile stresses to Cu coating/steel substrates (courtesy of NWMO and Exova).

KMCu–HC) used in the production of the cold spray coatings were supplied by the National Research Council (NRC), Boucherville, Québec. Nominally, the Cu particles have a diameter between 5 and 20 μ m and contain a low oxygen content of 0.190 \pm 0.004%. To facilitate their characterization, these powders were stirred and mounted in a mixture of carbon fibres and Struers EpoFix[®]resin and hardener to form a black solidified epoxy disc with a diameter of 2.5 cm. Carbon fibres were used to mitigate charging caused by the electron beam used in EBSD measurements. One face of the disc surface was prepared for EBSD measurements using the procedure described in Section 2.4.

2.2. Cu cold spray coatings

Cu cold spray coatings (3 mm (0.118") thick) on 6.35 mm $(\frac{1}{4}")$

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