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CORROSION OF COPPER-COATED STEEL HIGH LEVEL NUCLEAR WASTE CONTAINERS UNDER PERMANENT DISPOSAL CONDITIONS

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ABSTRACT

The corrosion of high level nuclear waste containers under permanent disposal conditions can occur via a number of processes which change in importance as the environment within a deep geologic repository evolves from oxic to anoxic. The container design involves a copper coating on a steel vessel. The characteristics of these coatings are being assessed using standard analytical methods, including microscopy and electron backscatter diffraction. The corrosion processes on the surface of the coatings and at through-coating defects are also being investigated electrochemically. Although minor differences exist, the surface corrosion of cold sprayed and electrodeposited coatings generally exhibited similar corrosion behaviour to standard wrought copper. Corrosion at the base of a simulated through-coating defect in a cold-sprayed coating was found to proceed via galvanic coupling to oxygen reduction on the coating surface. Besides accumulating damage at the base of the defect, corrosion propagated along the copper/steel interface, most likely a consequence of the damage inflicted during the deposition process.

1. INTRODUCTION

Like many nuclear nations, Canada has been investigating geological disposal of nuclear waste since the early 1970s, as this approach offers the best passive safety system for permanent disposal. The internationally accepted design of a deep geologic repository (DGR) involves disposal of the used fuel at a depth of ≥500 m in a suitably dense intact rock. To ensure containment, the nuclear fuel will be sealed in a corrosion resistant used fuel container (UFC) capable of withstanding the anticipated hydrostatic, lithostatic and glaciation loads. To provide an additional barrier, the UFC will be surrounded by compacted bentonite clay which swells on contact with moisture to tightly seal the system and provide a local environment in which only very little chemical diffusion can occur. A schematic representation of the proposed Canadian DGR is shown in Fig. 1.

The original Canadian UFC was very similar to the Scandanavian KBS-3 container and designed to contain 260 CANDU fuel bundles, which are ~50 cm in length and 10 cm in diameter (Fig. 1). These dual-walled containers are designed with an inner iron (or steel) bolted vessel to provide strength, and a separately fabricated 25-50 mm-thick Cu outer shell corrosion barrier. The dimensions of this shell are dictated by the container fuel capacity and manufacturing requirements, a significant wall thickness being necessary to provide sufficient stiffness to avoid warping of a shell this size. However, conservative estimates [1] suggest only a small fraction of this available thickness will be required as a corrosion allowance (≤1.3 mm over 10^6 years) even in groundwater containing unexpectedly high concentrations of sulphide (i.e., 3 ppm), which is known to corrode Cu.

Beginning in 2011, the Nuclear Waste Management Organization (NWMO) initiated a program to incorporate Scandinavian efforts to improve container design and to take advantage of recent improvements in manufacturing technologies and the unique small size of the CANDU fuel bundle. Presently, two conceptual designs are under consideration, Fig. 2. The 288 CANDU bundle Mark 1 retains the original Canadian design but has been altered to more closely align with the Scandinavian design with respect to geometry, enabling the transfer of technology between the two programs. However, manufacturing challenges remain with this design. To overcome these issues, while taking advantage of the CANDU bundle dimensions, the 48 bundle Mark 2 design, which utilizes an integral Cu coating on a fully welded steel container, is

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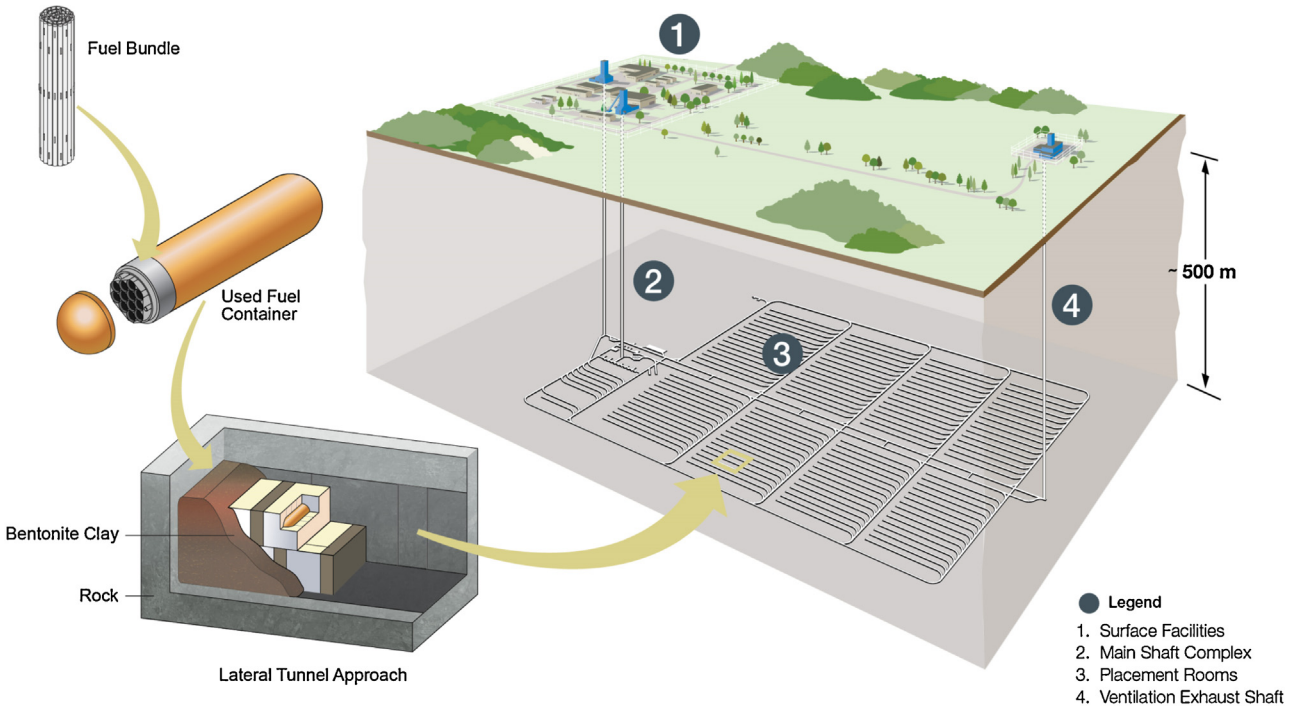

Fig. 1. Schematic representation of the proposed Canadian Deep Geologic Repository (DGR).

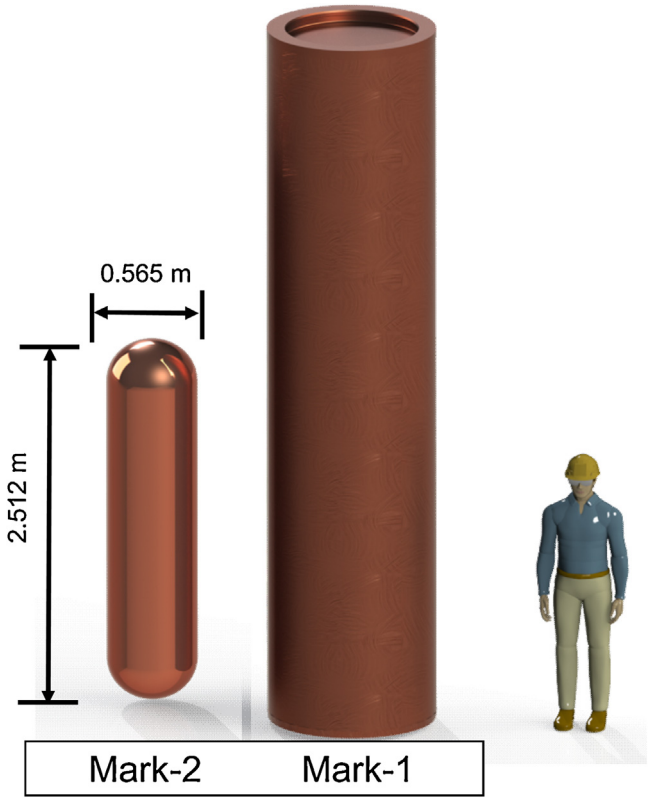

Fig. 2. The two conceptual container designs under consideration for the disposal of Canadian high level nuclear waste.

being considered. As depicted in Fig. 2, the Mark 2 is also substantially shorter (and lighter) than the Mark 1 which offers flexibility in handling. The use of commercial processes such as cold spray coating and electrodeposition are being considered [2,3] and since the required corrosion allowance is small, the corrosion performance of coatings with a thickness of a few mm is being evaluated [4].

Any chosen repository location must obviously be geologically stable and possess clearly identifiable advantageous features, which must not be permanently damaged by the thermal/chemical transient imposed by the excavation and waste emplacement process. Additionally, the evolution of the repository from the initially disturbed to the final long term condition must be predictable with reasonable certainty. While total corrosion is expected to be very small in a repository, the conditions within a Canadian repository will evolve from an initially aggressive initial state to an eventually benign state, Fig. 3. Upon repository closure, the temperature of the container surface will rise due to the heat produced by the radioactive decay of the spent fuel waste form in the container. It will also be oxidizing due to a combination of a small amount of oxygen, introduced during the period of repository excavation and emplacement of the waste, and the potential production of radiolytic oxidants due to gamma radiolysis of the environment. However, the rise in temperature will produce a low humidity environment in the vicinity of the container and negligible corrosion would be expected despite the oxidizing conditions.

Subsequently, moisture will return as the containers cool until full resaturation of the local environment is achieved. The period required for resaturation (i.e., the establishment of a relative humidity (RH) = 100%) is uncertain but expected to be between 20 and 100 years for a crystalline repository (as indicated in Fig. 3) but much longer for a sedimentary clay repository (i.e., >1000 years). Also, it is possible resaturation will occur non-homogeneously and rewetting of the surface will not be uniform. As resaturation progresses, trapped O_2 will be consumed by Cu corrosion, reactions with minerals and organic material in the clay, and biological processes. Consequently, repository conditions will evolve from