



An application of stochastic modeling to pitting of Spent Nuclear Fuel canisters

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ABSTRACT

Chloride induced stress corrosion cracking (CISCC) is one of the main factors affecting the integrity of used nuclear fuel in dry storage canisters, especially at coastal sites. CISCC has complex interactions associated with environment, stress and materials properties. This paper is focused on the development of probability distribution functions for maximum pit depth based on experimental data created at Lawrence Livermore National Laboratory (LLNL) that have not been fully analyzed before. The LLNL report outlined the key variables of pit initiation, termination and growth rate that may lead to CISCC, with the final aim of prediction of canister life and optimization of the time interval inspection of the canisters. The key parameters characterizing the probability distributions of pits at each stage depend on environment, material susceptibility conditions, and residual stress intensity. A basic stochastic approach to estimate probability distributions based on the median and maximum pit depths observed in experimental data succeeds in reproducing the experimental results, and can be used to estimate the distribution of maximum depth at future times.

1. Introduction and motivation

Currently, Spent Nuclear Fuel (SNF) is stored in two types of environmental storage conditions: 1) submerged in water in pools at reactor facilities, and 2) in dry storage at Independent Spent Fuel Storage Installations (ISFSIs), adjacent to reactor facilities. Generally after a few years of cooling time, SNF is removed from the water pool and transferred to helium-filled stainless steel canisters in passively ventilated dry storage systems.

Decisions have been made not to pursue Yucca Mountain as a long-term geologic repository storage facility for SNF and to cancel the construction of a reprocessing facility in the 1980s due to proliferation risks. As a result, interim dry storage facilities for long-term storage have been vital for the immediate future of nuclear energy technology. The Nuclear Regulatory Commission (NRC) and DOE have identified potential deterioration mechanisms for steel canisters containing the SNF in dry storage that require detailed research and investigation. This will have an impact on the performance of long-term interim storage under the normal and extreme environmental conditions experienced during the duration of this storage. The prediction and monitoring of canister corrosion processes while in storage can provide important information for the assessment of interim storage performance and the safety to the public.

One of the primary concerns with respect to the long-term

performance of storage casks is the potential for corrosion initiation due to deliquescence of salts deposited on the canister surface as aerosols; in regions of high residual weld stresses this may lead to localized stress corrosion cracking (SCC). Dust and aerosols in the air that are drawn through ventilation openings in the overpacks of passively-ventilated dry canister storage systems may be deposited on the stainless steel canister outer surfaces. Under these conditions, localized corrosion attack can occur. Chloride-induced stress corrosion cracking (CISCC) of welded zones is of special concern, as it is a well-documented mode of attack for austenitic stainless steels (including 304SS and 316SS) in marine environments (Kain, 1990), and many independent spent fuel storage installations (ISFSIs) are located in coastal areas. Recent canister inspections (EPRI, 2014; Bryan and Enos, 2014) have shown that chloride salts are present on the surface of in-service canisters in near-marine settings. However, canister surface inspections of sufficient resolution to detect SCC have never been carried out, because access to the canister surfaces through vents in the overpacks is extremely limited, and high radiation fields make removal of the canisters from the overpacks undesirable.

2. CISCC in dry storage canisters

SCC has been a research topic for many years. However, only a fraction of the large body of available research is relevant to the unique

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conditions of dry storage canisters. The time scale from pit initiation and growth, transition to crack, and crack growth rate is still the subject of much research. The current knowledge related to dry storage canisters is summarized based on the three necessary conditions for CISC (Environment, Susceptible Material, and Residual Stress).

2.1. Environmental conditions

In order for an environment to be aggressive, two conditions must be met: a corrosive chemical species must be present and aqueous conditions must exist. Other aggressive species may be present as well (e.g., high atmospheric concentrations of SO_2). This paper focuses on chloride species; that is, only CISC is being considered in this study. An aqueous condition can occur as the canister cools, as salts deposited as aerosols on the canister surface deliquesce (absorb water) to form brines. The quantity of deposited chlorine on canister surface is an important factor of CISC. Estimates of the minimum amount of chloride to support SCC include 0.3 g/m^2 (Shirai et al., 2011); 0.1 g/m^2 (Albores-Silva et al. 2011); and 0.056 g/m^2 (NRC 2014). Other authors have reported CISC at even lower salt loads, from 0.02 to 0.005 g/m^2 (Tokiwa et al., 1985; Taylor, 1994; Fairweather et al., 2008). Some studies have indicated that the salt surface load also has an impact on crack growth rate, as it affects the current-carrying capacity of the brine layer and the ability of the cathode, outside of the pit, to support corrosion at the anode, within the pit. This approach has been proposed for estimating maximum pitting penetration depths in several recent papers (for example, Chen and Kelly, 2010; Woldemedhin and Kelly, 2014; Krouse et al., 2014). To assess whether chloride is present on the canister surface requires knowledge of: (1) the composition of deposited salts and amount of chloride being deposited; and (2) the relative rates of chloride deposition versus chloride loss (e.g., via acid degassing) through time.

2.2. Susceptible material

Austenitic stainless steel is the regular material used in canister of dry storage casks, including Types 304, 304L, 316, and 316L, with the predominant material being Type 304 stainless steel. These materials are susceptible to SCC in aggressive environments. It occurs readily in experimental tests with deliquesced sea-salts (e.g., Nakayama, 2006; Tani et al., 2009; Mintz et al., 2012; Prosek et al., 2009, 2014), and has also been observed in near-marine ambient temperature field tests and industrial sites (Kain, 1990; Kosaki, 2008; Hayashibara et al., 2008; Nakayama and Sakakibara, 2013; Cook et al., 2011). However, the degree of susceptibility is a function of several factors, including the degree of sensitization, the degree of cold work, the presence of iron contamination on the metal surface, and the surface finish (Parrott and Pitts, 2011). The degree of sensitization is the most important parameter. However, when austenitic stainless steel is welded, the weld metal itself is melted and does not sensitize, but the Heat Affected Zone (HAZ) near the weld will become sensitized. Sensitization means that Chromium atoms in the austenite grains diffuse into nearby grain boundaries and combine with carbon to form chrome-rich carbides.

2.3. Tensile stress

The third condition for a CISC to grow is a tensile stress. The most suspected area in canister is a residual stress in the welding zone (in several studies, it is assumed that stresses imparted by cold working are too low to support SCC). There are no direct measurements of residual stresses associated with typical SNF dry storage casks in the welds zones. To assess the residual stresses in a full-diameter cylindrical canister the mockup should be buildup. The NRC recently reported an analysis of weld residual stress employing a 2D sequentially coupled thermal-structural analysis (Nuclear Regulatory Commission, 2013).

3. Pit initiation and growth

In most of the models, pitting initiation is based on nucleation-type theory in conjunction with the statistical methods used to describe rare-event processes when conditions for SCC are met (Turnbull et al., 2006). Two sources of information are available with respect to pit occurrence:

- Accelerated laboratory experiments: a high temperature and aggressive environment are used leading to fast (within days) occurrence of pits, although fast pitting, especially for heavily sensitized materials, is reported in the open literature (The data analyzed in this paper is of this type).
- Observation at ambient temperatures: these observations generally lead to slower occurrence of pits.

However, very few studies have been reported in the literature regarding the environment of interest for storage canister surfaces, and what is usually re-reported is the maximum pit depth observed rather than the evolution of pit depth as a function of time, rendering the model by Turnbull and Zhou (2004, 2006) difficult to parameterize for the present study. Rather, a statistical approach for the formation of stable pits is used in most models and in also this paper. One of the challenging tasks from a statistical point of view is to perform experimental work to parameterize a pitting initiation and growth model, to better estimate incubation times prior to the transition of pits to stress corrosion cracks. The CISC incubation times are a major uncertainty in predictive models for SCC.

3.1. Pit growth model

A typical pitting growth model is described by Turnbull et al. (2006). It has the form:

$$x_{pit} = \alpha_{pit} t^{\beta_{pit}}$$

where x_{pit} is pit depth, t is time after initiation, α_{pit} is a scaling factor, and the exponent β_{pit} is, in part, a function of the pit geometry and determines the shape of the growth curve with time for various environmental and materials conditions. Parameters α_{pit} and β_{pit} are generally determined experimentally for a given system and environmental conditions. Pitting corrosion rates are strongly affected by temperature, pH, (Relative Humidity) RH, and chloride concentration of the bulk solution (Shoji and Ohnaka, 1989; Ernst and Newman, 2002; Lu et al., 2008; Chen and Kelly, 2010; Asaduzzaman et al., 2011; Saadawy, 2012; Cook et al., 2011; Davenport et al., 2014; Krouse et al., 2014; Vagbharathi and Gopalakrishnan, 2014), so experimentally determined rates would only be relevant for specific system. In reality the deliquesced sea-salts, undergo randomly variable periodic evaporation and dry out at canister surface. Pits growth rates are a function of many different parameters and can be expressed in the following general form:

$$dx_{pit} = \alpha_{pit} f(T) f(K) f(R_s) f([Cl^-]) f(m_{Cl}) f(pH) f(O) f(\sigma_{ys}) \dots dt$$

where α_{pit} is the pit growth amplitude factor (at a fixed reference set of conditions), which can be modified by many other factors, including (1) material property factors such as the stress intensity factor (K), degree of sensitization (R_s), and yield stress (σ_{ys}), and (2) environmental factors such as temperature (T), chloride concentration ($[Cl^-]$), oxygen concentration (O), the mass of chloride per unit surface area (m_{Cl}), and the solution pH. Most of these variables fluctuate randomly. Therefore pit initiation and growth rate are subjected to random mechanisms. The authors are not aware of any theoretical physical models that can provide adequate prediction for various conditions or pit initiation and growth rates as stated above.

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